THE ALPHA-GAMMA ANGULAR CORRELATION IN THE DECAY OF RADIOTHORIUM

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ABSTRACT

A measurement is reported of the correlation between the directions of emission of the alpha rays and succeeding gamma rays in the decay of radiothorium to Thorium X. In this decay alpha rays having an energy of 5.33 Mev are followed by two gamma rays whose energies have been reported to be 82.3 and 86.8 Kev. Both the alpha and gamma radiations were detected by suitably designed scintillation counters. The observed angular correlation for the two gamma rays together was rather sharp:

$$W(\theta) = 1 + 6.90\cos^2\theta - 7.07\cos^4\theta$$

(here θ is the angle between alpha and gamma rays.) With the help of absorbers, it was also possible to measure the correlation between the alpha particles and each gamma ray separately.

Attempts have been unsuccessful to explain the observed correlations with a simple decay scheme that is consistent with all the information available on the decay of radiothorium. Strong arguments can be advanced, however, to show that both gamma rays are electric quadrupole. The implications of this result and the observed correlations on possible decay schemes for radiothorium are discussed.

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SUMMARY

The description of excited states of nuclei involves the specification of two types of information, - the energies of the excited states and the spins. Both must be provided by studies of the radiations between nuclear states. The measurement of the energies, relative intensities, and other properties of nuclear radiations is consequently one of the important branches of experimental nuclear physics.

The correct assignment of energy levels and their spins to a nucleus must be consistent with a large amount of data. The consistency of some of the data with a given level scheme involves basic theoretical assumptions about the nuclear radiations. For example, whether or not an observed conversion coefficient fits a given level assignment, depends on the theoretical dependence of conversion coefficients on energy and spin differences and other nuclear parameters.

It is therefore desirable to confront a level assignment with an overabundance of data. A consistent fit would imply the validity of the basic theoretical assumptions involved in the fit.

In the past it has been harder to get spin data than energies for excited nuclear states. Energy differences of levels can be measured directly but spin differences can only be inferred. Recently a powerful technique has been brought

to bear on the spin problem, the technique of measuring the angular correlation of successive nuclear radiations. In many cases, such a measurement can provide a unique spin assignment to the three nuclear levels involved in the radiations.

In the present experiment the angular correlation between alpha particles emitted by Radiothorium and the gamma rays which follow was measured. Radiothorium was chosen for the experiment because a fair amount of information was available on its decay scheme.

Radiothorium decays to Thorium X by the emission of an alpha particle. About one fortieth of the disintegrations are accompanied by gamma radiation, and this radiation has been reported to consist of quanta of energies of 83.2 and 86.8 Kev. Most of the alphas have an energy of 5.42 Mev. but about one third of them are in a group 86.7 Kev less energetic than the main group. There is no other group, implying that the two gamma rays do not occur in cascade. This data, with some measurements of the conversion coefficients represents all the experimental information that was available on the decay of radiothorium.

In the present experiment a thin source of radiothorium, chemically separated from its decay products, was mounted in the center of an evacuated cylindrical chamber on a thin

foil of polystyrene. The alpha particles were detected by a scintillation counter using a thin anthracene crystal which was fixed relative to the source. The source and alpha counter could be rotated around an axis through the center of the chamber. The gamma detector was a stationary NaI scintillation counter off to one side of the chamber.

After pulse-height selection the alpha and gamma pulses were fed to a coincidence circuit with a one-quarter microsecond resolving time. Coincidence rates were recorded as a function of the angle between the counters. A run was permitted to last no longer than ten hours in order to keep possible correlations due to the radiothorium decay products from creeping into the data.

The 83.3 and 86.8 Kev gamma rays of ThX were detected with equal efficiencies in the gamma counter and the angular correlation observed was the weighted average of the individual correlations involving each ray. Measured to a statistical accuracy of two percent, this correlation was

$$1 + 6.90\cos^2\theta - 7.07\cos^4\theta.$$

A thallium absorber of thickness 0.5 $\frac{gm}{cm^2}$ placed in front of the gamma counter would reduce the intensity of an 83.3 Kev gamma to one third, and practically completely remove 86.8 Kev radiation because the K absorption edge of thallium is at 86.1 Kev. The correlation measured with

thallium covering the gamma counter was therefore attributed to the 83.3 Kev gamma ray alone and was

$$1 + 6.81\cos^2\theta - 7.33\cos^4\theta$$

with statistical errors of about 8%.

The correlation absorbed in the thallium was

$$1 + 7.2\cos^2\theta + 6.2\cos^4\theta$$

with statistical errors of 30%.

Before summarizing the possible interpretations of these results, it should be remarked that it was possible to confirm a number of aspects of the radiothorium decay scheme in the course of the present experiment.

1) The pulse heights of the radiothorium gammas corresponded to energies between 80 and 90 Kev and absorption in thallium, lead, and gold, showed that there were at least two such gamma rays, one on each side of the Thallium K edge.

2) No coincidences between the gamma rays could be found, showing that they do not occur in cascade.

3) Gamma radiation was found to be emitted in only about one of every forty alpha emissions.

4) Both gamma rays have lifetimes less than 0.5×10^{-6} sec. Otherwise they would have not been observed in coincidence with the alpha particles.

5) The gamma rays must be at least quadrupole and the states from which they originate must have a spin of at least two. This follows from the existence of large $\cos^4\theta$ terms in the angular correlations.

The observed correlations do not fit into a simple picture of the levels involved in the decay of radiothorium that is consistent with other requirements. The decay starts in the ground state of radiothorium and presumably ends in the ground state of ThX. Both these nuclei have even atomic weights and even atomic numbers and, therefore, would be expected to have a spin of zero in their ground states. Either the excited state or the ground state of ThX must be a close doublet in order to account for the two gamma rays. The observed correlations are inconsistent with the assignment of zero spins no matter where the doublet is located.

If it is insisted that the ground state spins of RdTh and ThX are zero, then at least one of the following must be true.

1) The assumed decay scheme is too simple and more than four levels are really involved.

2) The observed correlation differs from the simple correlation that can be computed from the spins because appreciable spin reorientation of the excited state of ThX takes place before gamma emission.

3) The observed correlation suffers from systematic experimental errors that are far in excess of the probable errors involved in the measurements.

Consideration of these alternatives indicates that none seems to be particularly likely to account for the trouble in trying to interpret the observed correlations.

This report concludes with a discussion of the possible attempts at obtaining a fit of the correlations to the level schemes which are consistent with other ex-

perimental evidence. This discussion includes a fairly detailed display of both the experimental evidence that is to be correlated and the theoretical assumptions used in their correlation. On the basis of the discussion some suggestions are made for future investigations of the radiothorium decay scheme.

CHAPTER I

Introduction

In the past few years a technique has developed which promises to increase significantly the knowledge of one of the most important quantized properties of nuclei. It has become possible, in certain cases, to measure the correlation between the directions of emission of successive radiations from an atomic nucleus. It often happens that two radiations occur in such quick sequence that the nucleus has negligible chance of being disturbed by fields of neighboring atoms between emissions. In these cases measurement of the relative probability that the second radiation be sent out at a specified angle to the direction of the first generally gives information about the angular momenta involved in the emission process.

The first published reference to the possibility that such an angular correlation might exist appears to have been made by Dunworth in 1940. The idea was suggested to him by his experimental studies on the counting of coincidences between nearly simultaneous nuclear events, (1.1). Hamilton developed the basic theory in the same year, (1.2).

During the next five years there were a number of attempts to measure angular correlations between gamma rays.* These failed to give definite indications of directional effects as large as predicted mainly because the geiger counters used had such low intrinsic efficiencies that apparatus would not remain stable long enough to get significant data.** It is hard to explain why experimenters persisted so long in attempting measurements which constrained them to extremely low counting efficiencies. There should have been strong temptation to do an alpha-gamma or beta-gamma correlation experiment, if only to see what might happen, because a gain in counting rate of a factor of a hundred over the gamma-gamma case could have been realized. An example of a specific case where a correlation should have been measurable with the electronics of ten years ago is the alpha decay of ThC which is followed by a 40 kev gamma ray. A geiger counter can be made to have a fairly high efficiency for this energy and the gamma was studied by Ellis in 1932 and identified as quadrupole by Ellis and Mott in 1933, (1.6). Successful gamma-gamma angular correlation experiments became possible with the advent of scintillation counters and were reported by Brady and Deutsch in 1947, (1.7).

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* See for example 1.3 and 1.4.

** A way of balancing out drifts is suggested in 1.5.

The correlation method finds its application to a known or probable decay scheme. Angular momenta are assumed for the nucleus before, between and after the emissions and angular momentum properties consistent with conservation of parity are assigned to the emitted particles. The angular correlation is computed and compared with experiment. In most situations there are other conditions than parity which severely limit the possible angular momenta. These constraints might be known gamma ray lifetimes, internal conversion coefficients, shape of beta ray spectra and the like. There is a fair degree of uniqueness to correlation functions so that it is often possible to determine a number of unknowns from a single experiment. Taken in conjunction with other information the measurement of an angular correlation may give information about any of the parameters needed to calculate the correlation. The next chapter will say in detail what these are, but it is worth remarking here that they may include nuclear matrix elements.

The experiment described here is an application of the angular correlation technique to the decay of radiothorium. That nucleus is one of the even-even alpha emitters of the thorium series which have been generally assumed to have ground states without angular momentum.

Only radiothorium and ThX of these six nuclei have appreciable alpha fine structure. Information on the spins of excited states from which gamma rays are emitted has been unreliable because of the present status of internal conversion calculations. Spins for these states have been assigned mainly by application of a completely inadequate theory of alpha decay, (1.8). Measurement of the angular correlation in the disintegration of radiothorium is essentially an intensity problem, there being only a fortieth as many gamma rays as alpha particles. Nevertheless, it is well worth attempting because a successful determination of the correlation would be a completely independent method of measuring spins and a check on past assumptions of zero spin for the ground states of radiothorium and ThX. It is possible that a similar experiment on ThX would be less difficult; however the gamma radiation to be expected from the known alpha branching has not yet been observed.

CHAPTER II

Angular Correlation Theory

Section 2.1. Introduction

It is the purpose of this chapter to present enough angular correlation theory for interpretation of data from the radiothorium experiment. The treatment is, therefore, not particularly general.

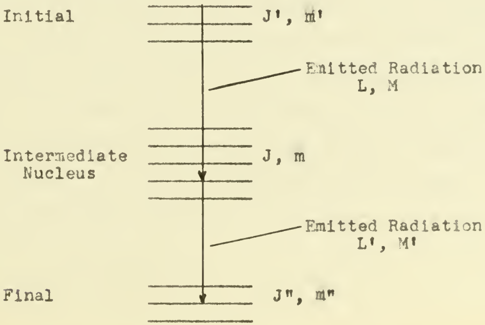
The existence of a correlation between the directions of emission of successive nuclear radiations is essentially a quantum mechanical phenomenon. It stems from the special role that angular momentum has in quantum mechanics, in combination with the principle of conservation of angular momentum for an isolated system. Only weak and unilluminating classical analogies can be made. A semi-quantum mechanical picture based on the vector model and the uncertainty principle (2.1) has some fortuitous success for simple cases, but cannot be applied generally.

Section 2.2. Elementary Theory

A naive attempt at the calculation of an angular correlation might treat the process as the causal compounding of two separate transitions. This would, in general, be wrong because the assignment of probabilities

to unobserved intermediate processes instead of probability amplitudes neglects interference between ways the overall process can occur. However, such a derivation turns out to be correct when applied with a simple restriction and is therefore worth presenting as a mnemonic device.

Consider then an isolated initial nucleus with angular momentum quantum number, J', having as Z component relative to an arbitrary axis one of $2J' + 1$ possible values, say m'. Emission of the first particle leaves the intermediate nucleus in the state J, m, and after the second particle comes out the nucleus is in the state J'', m''. The figure below shows the two transitions schematically. Each nuclear energy level with angular momentum J consists of a set of $2J + 1$ degenerate levels with differing Z components of angular momentum, often called magnetic sub-states.



The probability that the first particle be emitted at an angle θ_1 to the Z axis in the transition $J', m' \rightarrow J, m$ will depend on the substates involved and the nature of the radiation, and may be called $P_{m,M}(\theta_1)$. In the present view, the probability of observing two particles emitted at angles θ_1 and θ_2 with the final nucleus left in the state J'', m'' will be the product of the individual probabilities $P_{m,M}(\theta_1)P_{m'',M'}(\theta_2)$. The probability of forming the final nucleus in all substates following the transition J', $m' \rightarrow J$, m will be $\sum_{m''} P_{m,M}(\theta_1)P_{m'',M'}(\theta_2)$ the radiations having been emitted at angles θ_1 and θ_2. Summing this expression over m' gives the total probability that the magnetic state m be involved in the emissions. A further sum over m gives the probability of emitting the two radiations at the specified angles in the transformation from J' to J''. This is the desired angular correlation $W(\theta_1, \theta_2) = \sum_m \sum_{m'} \sum_{m''} P_{m,M}(\theta_1)P_{m'',M'}(\theta_2)$. This equation is immediately to be suspected because it asserts that the angular correlation depends on the angles θ_1 and θ_2 relative to an arbitrary Z axis. So long as the specification of an axis does not introduce a potential energy into the problem there is no physical basis for expecting the correlation to depend on whatever axis is picked to describe the angular features of the decay. W should,

therefore, be a function only of $|\theta_1 - \theta_2|$. It has been shown* that to correct the previous derivation it is only necessary to replace the arbitrary axis by one chosen along the direction of emission of either particle. Suppose the first radiation defines the Z axis. Then $\theta_1 = 0$ and θ_2 may be called θ giving

$$W(\theta) = \sum_{m,}\sum_{M',}\sum_{m''} P_{m,M}(0)P_{m'',M'}(\theta). \quad \text{Eq. } (2.1).$$

This expression, or the corresponding one for the case when the second emission direction is the axis, can be used for the calculation of correlation functions. Before illustrating such a calculation, its range of validity will be indicated by stating the assumptions implicit in the rigorous derivation of $W(\theta)$. These are that all three levels are discrete and of definite parity; the emitted radiations are either gamma rays or particles with non-relativistic velocity**; the orientation of the angular momentum of the initial nucleus is random and finally that the angular momentum vector of the intermediate nucleus is not reoriented in the lapse of time before the second radiation is emitted. In the radiothorium decay these conditions are satisfied, except possibly the last, and even here it can be made quite plausible that reorientation is negligible.

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* 2.2, 2.3, 2.4.

** For a discussion of angular correlations involving β particles, see 2.5.

It is worth noting here that Spiers recent derivation (2.4) of Eq. 2.1 by second order time dependent perturbation theory is particularly readable.

Section 2.3. Discussion of Simple Correlations

In the emission of radiation of total angular momentum L by an isolated nucleus with angular momentum J' to form a nucleus with quantum number J, the angular momentum of final nucleus plus emitted particle must add vectorially to that of the original nucleus. There are, in general, a number of values of L consistent with conservation of angular momentum. If J and J' are expressed in units of $\hbar$ these range by integral steps from $|J' - J|$ to $J' + J$. In many cases, however, only a single value of L has appreciable probability of emission. This section is concerned with simple correlations where there is only one value for each transition, namely L for the first and L' for the second.

In case a unique L is involved in a transition $J', m' \longrightarrow J, m$ the probability $P_{m,M}(\theta)$ can always be written*, $P_{m,M}(\theta) = \alpha\, G^{J',J,L}_{m,\ M} F^L_M(\theta)$.

In this relation, $G^{J',J,L}_{m,\ M}$ is the square of the well-known Clebsch-Gordan coefficient for the quantum mechanical addition of angular momenta, $F^L_M(\theta)$ is the angular distri-

* See 2.3 for proof and a general discussion of correlations not involving mixtures of different angular momenta for the emitted radiations.

bution of the emitted radiation and α is a proportionality factor not involving m, M or m'. With this decomposition of the transition probabilities, Eq. 2.1 becomes $W(\theta) = \alpha\beta \sum_{m,m'}\sum_{,m''} g^{J',J,L}_{m,M} f^{L}_{M}(0) G^{J,J'',L'}_{m'',M'} F^{L'}_{M'}(\theta)$.

Here small Latin letters refer to the first transition and capitals to the second. Since α and β, which is the factor corresponding to α for the second transition, do not involve magnetic quantum numbers they can come outside the sums. They may be omitted entirely if the convention is made that proportionality will be counted equality. This will be done, usually without comment, because the interest in $W(\theta)$ lies in its relative value. then $W(\theta) = \sum_{m,m'}\sum_{,m''} g^{J',J,L}_{m,M} f^{L}_{M}(0)\ G^{J,J'',L'}_{m'',M'} F^{L'}_{M'}(\theta)$.

Since the Z component of angular momentum must be conserved for each transition, $m' = m + M$ and $m = m'' + M'$. It is, therefore, necessary to specify only two magnetic quantum numbers per transition and $W(\theta)$ may be conveniently rewritten,

$$W(\theta) = \sum_{m,M,M'} g^{J',J,L}_{m,M} f^{L}_{M}(0)\ G^{J,J'',L'}_{m-M',M'} F^{L'}_{M'}(\theta) \qquad \text{Eq. (2.2)}$$

For successive emission of alpha and gamma rays with Z axis chosen along the direction of motion of the alpha particle, there can be no change in the Z component of angular momentum in the first transition. This is because an alpha particle has no intrinsic spin, hence no Z component

of angular momentum along its direction of flight. The impossibility of a change in magnetic quantum number is recognized in the property of the alpha particle angular distributions, $f_M^L(0) = (\text{constant})\ \delta_{m',m}$.

$W(\theta)$ for an alpha-gamma angular correlation can therefore be reduced to the double sum,

$$W(\theta) = \sum_{m=m'} g_{m,0}^{J',J,L} \sum_{M'} G_{m-M',M'}^{J,J'',L'} F_{M'}^{L'} \qquad \text{Eq. (2.3)}$$

This is the form used in calculations. The alpha particle angular distributions, which are just proportional to squares of the associated Legendre polynomials of order LM, have dropped out. If the gamma ray had defined the Z axis they would have appeared in place of the gamma distributions. These latter have been derived by the correspondence principle for arbitrary angular momentum (multiple order). (2.6). They are needed only for orders one and two for this experiment, and are:

$$L = 1:\quad F_0^1 = 1 - \cos^2\theta$$

$$\text{Dipole Radiation}\quad F_{\pm 1}^1 = \tfrac{1}{2}(1 + \cos^2\theta)$$

$$L = 2:\quad F_0^2 = 6(\cos^2\theta - \cos^4\theta)$$

$$\text{Quadrupole Radiation}\quad F_{\pm 1}^2 = 1 - 3\cos^2\theta + 4\cos^4\theta$$

$$F_{\pm 2}^2 = 1 - \cos^4\theta$$

The Clebsch-Gordan coefficients $(G^{J',J,L}_{m,M})^{\frac{1}{2}}$ are tabulated for L = 1, 2 (2.7). For higher L they may be worked out.*

Section 2.4. Calculation of a Simple Correlation

A specific alpha-gamma correlation calculation will now be illustrated. Let the initial and final states have spins of unity while the intermediate nucleus has spin three. The alpha and gamma rays are both to have angular momentum two.

Then $J' = J'' = 1$, $J = 3$, $L = L' = 2$.

$$W(\theta) = \sum_{m=-1}^{1} g^{1,3,2}_{m,o} \sum_{M'=-2}^{2} G^{3,1,2}_{m-M',M'} F^2_{M'}(\theta)$$

$$W(\theta) = \sum_m g^{1,3,2}_{m,o} \left\{ G^{3,1,2}_{m,o} F^2_o + (G^{3,1,2}_{m-1,1} + G^{3,1,2}_{m+1,-1}) F^2_1 + (G^{3,1,2}_{m-2,2} + G^{3,1,2}_{m+2,-2}) F^2_2 \right\}$$

$$g^{1,3,2}_{m,o} = \frac{3(3-m)(2-m)(3+m)(2+m)}{4.5.3.7} \qquad \text{(From pg. 77, 2.7)}$$

$$= (9-m^2)(4-m^2) \qquad \text{(Factors dropped)}$$

$$G^{3,1,2}_{m,o} = \tfrac{3}{2}(3-m)(2-m)(3+m)(2+m) = \tfrac{3}{2}(9-m^2)(4-m^2)$$

$$G^{3,1,2}_{m-1,1} = (3-m)(3+m)(2+m)(1+m)$$

$$G^{3,1,2}_{m+1,-1} = (3-m)(2-m)(1-m)(3+m)$$

$$(G^{3,1,2}_{m-1,1} + G^{3,1,2}_{m+1,-1}) = 2(9-m^2)(2+m^2)$$

$$G^{3,1,2}_{m-2,2} = \tfrac{1}{4}m(1+m)(2+m)(3+m)$$

$$G^{3,1,2}_{m+2,-2} = -\tfrac{1}{4}m(1-m)(2-m)(3-m)$$

$$(G^{3,1,2}_{m-2,2} + G^{3,1,2}_{m+2,-2}) = \tfrac{1}{2}m^2(11+m^2)$$

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* Eq. 16, pg. 428, 2.8

$$W(\theta) = \sum_{m=-1}^{1} (9-m^2)(4-m^2) \Big[\tfrac{3}{2}(9-m^2)(4-m^2)F_o^2 +$$
$$2(9-m^2)(2+m^2)F_1^2 + \tfrac{1}{2}m^2(11+m^2)F_2^2 \Big]$$

This is symmetric in m so that $W(\theta) = W_{m=o} + 2W_{m=1}$

$$W_{m=o} = 9\cdot4(\tfrac{3}{2}\cdot9\cdot4F_o^2 + 9\cdot4F_1^2) = 6^4(\tfrac{3}{2}F_o^2 + F_1^2)$$

$$2W_{m=1} = 2\cdot8\cdot3(\tfrac{3}{2}\cdot8\cdot3F_o^2 + 2\cdot8\cdot3F_1^2 + 6F_2^2) = 6^2 \cdot 8(6F_o^2 + 8F_1^2 + F_2^2)$$

$$W(\theta) = 9(\tfrac{3}{2}F_o^2 + F_1^2) + 2(6F_o^2 + 8F_1^2 + F_2^2) = 51F_o^2 + 50F_1^2 + 4F_2^2$$

Substituting for the Fs gives,

$$W(\theta) = 54 + 156\cos^2\theta - 110\cos^4\theta = 27 + 78\cos^2\theta - 55\cos^4\theta \quad \text{Eq.(2.4)}$$

$$W(\theta) = 1 + \tfrac{26}{9}\cos^2\theta - \tfrac{55}{27}\cos^4\theta$$

Which is the angular correlation normalized to unity at 90°.

Falkoff and Uhlenbeck (2.3) give convenient cannonical forms for simple correlations where neither radiation has $L > 2$. For these cases $W(\theta)$ may therefore be checked independently with little effort.

Section 2.5. The Likelihood of Mixed Correlations

The possibility that the radiation in a transition between states with angular momenta J' and J will be emitted as a mixture of angular momentum values must be considered

when both J and J' are different from zero. Denoting the smaller angular momentum by J, there will be 2J + 1 possible values of L. The parity selection rule will permit either J or J + 1 of these. If more than a single L is permitted the transition will occur as a mixture unless the emission probabilities for all but one of the different angular momenta are negligible. For gamma radiation the probability of emission is a rapidly decreasing function of L so that mixed transitions are relatively uncommon. This is definitely not the case for alpha emission from heavy elements where emission probability varies slowly enough with L to make mixtures important whenever allowed by angular momentum and parity selection rules.

It is therefore obligatory to consider mixtures for the alpha radiation when attempting to interpret the measured radiothorium angular correlation. There is a possibility that the gamma radiation also be a mixture, or that a mixed gamma transition follow emission of an alpha particle, with single L value. Ling and Falkoff have discussed mixtures in gamma-gamma correlations (2.6) and their treatment can be applied to the alpha-gamma problem if the alpha part of the calculation is understood. The treatment of simple alpha emission has been illustrated. The

correlation calculation of interest is therefore the case of a mixed alpha transition followed by the gamma radiation of a single multipole.

Equation 2.1 is valid in general so the problem reduces to determining $P_{m,M'}(\theta)$, the angular distribution of alpha particles emitted in the transition $J',m' \rightarrow J,m$.

Section 2.6. A Single Alpha Transition

One of the possible treatments of alpha decay proceeds by first order time dependent perturbation theory. The alpha particle is initially bound by a potential slightly different from the actual one. At zero time a small interaction between particle and nucleus is turned on which results in the alpha feeling the true nuclear potential and having a probability of emission.

The time dependent solution of the Schrödinger equation, which has angular momentum quantum numbers J', m', is a linear combination of the obvious basis functions for the problem:

$$\Psi^{J'}_{m'}(t) = a_0(t)\ \psi'^{J'}_{m'} + \sum_L a_L(t) \sum_M C^{J',J,L}_{m=m'-M}\, \psi^{J}_{m'-M}(\xi)\, \phi^{L}_{M}(r,\theta,\varphi)$$

Here r,θ,φ are the usual polar coordinates expressing the location of the alpha particle relative to the product nucleus; ϕ^L_M is a free alpha particle wave function with the

indicated angular momentum properties; ξ denotes a set of nuclear coordinates; $\psi^{J}_{m}(\xi)$ is a wave function of the daughter nucleus; and $\psi'^{J'}_{m'}$ is the bound state wave function for the system prior to t=0. Because of conservation of angular momentum the sum contains only terms whose angular momentum and Z component equal the original values J',m', and is constructed using the Clebsch-Gordan coefficients, $C^{J',J,L}_{m,M}$. The probability amplitudes $a_L(t)$ are functions of energies and time multiplied by the matrix element between the states $\psi'^{J'}_{m'}$ and $\psi^{J}_{m}\phi^{L}_{M}$, and $a_o(t)$ is an exponentially decreasing function of time with $a_o(o) = 1$.

The probability density, $p_{m'}(r,\theta,t)$ of the emitted particle is obtained by integrating the square of the modulus of the sum in the previous expression over the coordinates of the product nucleus.

$$p_{m'}(r,\theta,t) = \int \left| \sum_L a_L(t) \sum_M C^{J',J,L}_{m'-M,M} \psi^{J}_{m'-M}(\xi) \phi^{L}_{M}(r,\theta,\varphi) \right|^2 d\xi$$

Since the nuclear wave functions having different m are orthogonal, the sum on M can be taken outside.

$$p_{m'}(r,\theta,t) = \sum_M \left| \sum_L a_L(t) C^{J',J,L}_{m'-M,M} \phi^{L}_{M} \right|^2$$

The probability density for emission with a given M is $p_{m'M}$ or since m+M=m' it can be written $p_{m,M}$.

$$p_{m,M}(r,\theta,t) = \left| \sum_L a_L(t) C^{J',J,L}_{m,M} \phi^{L}_{M} \right|^2$$

$P_{m,M}(\theta)$, the desired probability of alpha emission at an angle θ in the transition $J', m' \longrightarrow J, m$ will be had by evaluating $p_{m,M}(r,\theta,t)$ for fixed t and large constant r. The alpha particle wave function ϕ_M^L must have the asymptotic dependence of an outgoing wave with the angular properties of a solution of the time independent Schrödinger equation in the Coulomb field of the product nucleus.

Thus,* $\phi_M^L = \frac{f(r)}{r} e^{ikr} e^{i\delta_L} Y_M^L(\theta,\varphi)$

f_L is a function of r only, Y_M^L the spherical harmonic of order L,M, δ_L the phase shift due to the field of the residual nucleus and k is the magnitude of the propagation vector of the alpha particle. The appearance of k gives assurance that the perturbation method of calculation will yield the transition probability proportional to time which is observed. This is because k may vary slightly as allowed by the uncertainty principle so there are many final states of the system having energy very close to the initial energy** and separated into a few discrete groups according to their angular momentum characteristics.

Let $A_L = a_L \frac{f_L}{r} e^{ikr}$

then $P_{m,M}(\theta) = \left| \sum_L A_L e^{i\delta_L} C^{J',J,L}_{m,M} Y_M^L(\theta,\varphi) \right|^2$

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* Chapter IV, 2.9

** Chapter VIII, 2.9

Which is not a function of φ since Y_M^L contains $e^{im\varphi}$ which drops out when the modulus is taken.

Section 2.7. Example of a Correlation Calculation for Mixed Alpha Emission

Let $J' = J'' = 1$, $J = 3$, $L = L_\alpha = 2$ and 4, $L' = L_\gamma = 2$.

The direction of alpha emission is chosen as Z axis and the correlation function takes the form,

$$W(\theta) = \sum_{m=-1}^{+1} \sum_M P_{m,M}(0) \sum_{M'} G^{3,1,2}_{m-M',M'} F^2_{M'}(\theta)$$

The second transition is identical with the one in the simple correlation previously illustrated so that the sum on M' has already been evaluated.

$$2\sum_{M'} G^{3,1,2}_{m-M',M'} F^2_{M'}(\theta) = 3(9-m^2)(4-m^2)F_0^2 + 4(9-m^2)(2+m^2)F_1^2$$

$$+ m^2(11+m^2)F_2^2$$

$$P_{m,M}(\theta) = \left|\sum_L A_L e^{i\delta_L} C^{J',J,L}_{m,M} Y^L_M\right|^2 =$$

$$\left|A_2 e^{i\delta_2}(C^{1,3,2}_{m,M} Y^2_M + \frac{A_4}{A_2} e^{i(\delta_4-\delta_2)} C^{1,3,4}_{m,M} Y^4_M\right|^2$$

Let $\alpha = \frac{A_4}{A_2}$ and $\delta = \delta_4 - \delta_2$

Then $P_{m,M}(\theta) = (C^{1,3,2}_{m,M} Y^2_M)^2 + \alpha^2 (C^{1,3,4}_{m,M} Y^4_M)^2 +$

$$2\alpha \cos\delta\, C^{1,3,2}_{m,M} C^{1,3,4}_{m,M} Y^2_M Y^4_M$$

$$Y^L_M(0) = (\text{constant}) \sqrt{2L+1} \quad \delta_{M,0}$$

$$\therefore \sum_M P_{m,M}(0) = P_{m,0}(0) = 5(C^{1,3,2}_{m,0})^2 + 9\,\alpha^2(C^{1,3,4}_{m,0})^2 +$$
$$6\sqrt{5}\;\alpha\cos\delta\; C^{1,3,2}_{m,0}C^{1,3,4}_{m,0}$$

$C^{1,3,2}_{m,0}$ may be taken from page 77, 2.7, or computed from Eq. 16, 2.8. $C^{1,3,4}_{m,0}$ is computed.

They are,

$$C^{1,3,2}_{m,0} = \sqrt{\frac{3(3-m)(2-m)(3+m)(2+m)}{2.2.5.3.7}}$$

$$C^{1,3,4}_{m,0} = \frac{1}{6\cdot(84)^{1/2}}(7m^2 - 4)\sqrt{(4-m^2)(9-m^2)}$$

Substitution and reduction give,

$$W(\theta) = 2(27+78\cos^2\theta-55\cos^4\theta)+3\alpha^2(17+60\cos^2\theta-45\cos^4\theta)+$$
$$4\sqrt{3}\;\alpha\cos\delta\;(3-24\cos^2\theta+25\cos^4\theta)$$

The correlation consists of that for L=2, previously calculated, plus the correlation for L=4 weighted by the relative probability of emission with L=4, and an interference term. The quantity α is essentially a nuclear matrix element and unknown. It may be estimated reasonably well from the theory of alpha decay* as about 0.32 for radiothorium. It is clearly out of the question to ignore the existence of mixtures in calculating angular correlations involving alpha particles when amplitude ratios are of this order of magnitude.

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* For a modern treatment of alpha decay see 2.10.

Section 2.8. Properties of $W(\theta)$

Angular correlation functions have general properties which are important to recognize in a correlation experiment. The correlation can always be expressed as a sum of even powers of cosine; $W(\theta) = \sum_{i=0,1,\cdots} a_i \cos^{2i}\theta$. Eq. (2.5) Consequently, correlations should be symmetrical about $\theta = 0$ and $\theta = \pi/2$ which provides a check on experiment. The sum in Eq. 2.5 can be shown* not to extend beyond the smallest of the three quantum numbers L, J, L', where L and L' are the largest angular momenta involved in the first and second transition respectively. Consequently an isotropic correlation results from J = 0 and J = ½. An angular correlation which is observed to contain a $\cos^4\theta$ term, therefore, must involve angular momenta of the emitted radiations and intermediate nucleus at least equal to two.

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* See 2.3.

CHAPTER III

The Choice of Equipment

Section 3.1. The Experimental Problem

Radiothorium is an alpha emitter with a 1.9 year half life. The energy of the main group which includes about 72% of the alpha particles is 5.42 mev. The remaining alphas represent a disintegration energy 86.7 kev less than the main group (3.1). The first accurate measurement of the energy of the gamma radiation from the daughter, ThX, was made in 1941 by Surugue and Tsien (3.2) by magnetic analysis of L conversion electrons. Energies of 83.3 and 86.8 kev were found. These have been confirmed by Riou (3.3) who established that there are 1.8 quanta of 83.3 kev and 0.7 of 86.7 kev per hundred disintegrations.

Figure 3.1 shows the decay chain of radiothorium with half lives and disintegration energies. Other alpha emitters with significant fine structure are ThX where the five percent alpha branching could give rise to a gamma ray of about 250 kev, and ThC whose decay produces a 40 kev gamma having an intensity in equilibrium of three quanta per hundred RdTh disintegrations. The ThC decay also has less intense gamma rays of higher energy.

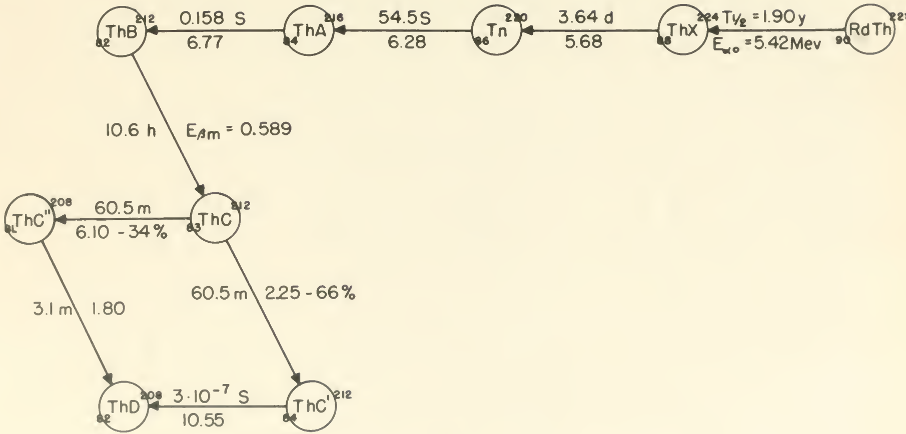


Figure 3.1

THE DECAY CHAIN OF RADIOTHORIUM

The beta emitters ThB, ThC and ThC" are responsible for a great deal of gamma radiation when the chain is in equilibrium with radiothorium. They also produce copious X rays in the neighborhood of 80 kev by internal conversion in the K shells of their daughters. Table 3.1 summarizes approximately the gamma radiation to be expected from a radiothorium source in secular equilibrium with its decay products.

The central problem of the experiment was the attainment of a reasonable true coincidence rate between alphas and gammas from the disintegration of radiothorium while excluding true coincidences due to the decay products. Immediate conditions on its solution are that the efficiencies of the alpha and gamma counters must be small, since it is desired to count only radiations having well defined directions, and that the accidental coincidence rate may not be excessive.

The solution of the problem is definitely non trivial but equally certainly not unique. The experiment could have been performed with alpha and gamma detectors different from those used here and with other electronics. However, the equipment actually used represents a reasonably logical choice as will be shown below.

TABLE 3.1*

Gamma Radiation from the Decay Products of Radiothorium in Secular Equilibrium. Intensities are Percent Quanta Per RdTh Disintegration

Transition		Gamma Ray Energy Mev.	Intensity %
RdTh	ThX	.0868	0.7
		.0833	1.8
ThX	Tn	.254	< 0.4
		.233	< 4.6
ThB	ThC	.24	46
ThC	ThC'	2.20	2
		1.80	4
		1.60	4
		1.35	2
		1.03	4
		0.80	10
		0.72	13
ThC"	ThD	2.62	34
		0.86	9
		0.58	24
		0.51	8

* Summary of data in National Bureau of Standards Circular 499. Sept. 1, 1950.

Section 3.2. Coincidence Counting in an Angular Correlation Measurement.*

Consider an angular correlation experiment on an alpha emitter, which might be radiothorium. In a fraction β, of the disintegrations a gamma ray follows its alpha in a time small compared with the resolving time, τ, of the coincidence circuit. The alpha counter has an intrinsic efficiency η_α for all alpha particles and is insensitive to gamma rays and correspondingly the gamma counter counts only gammas for all of which it has intrinsic efficiency η_γ. Let the source strength be D disintegrations per second and α and γ denote respectively the rates in the alpha and gamma counters. C and R are the true and random coincidence rates and g_α and g_γ the geometrical efficiencies of the counters.

Then

$$\begin{aligned} \alpha &= \eta_\alpha \; g_\alpha \; D \\ \gamma &= \eta_\gamma \; g_\gamma \; D \\ C &= \eta_\alpha \; g_\alpha \; \eta_\gamma \; g_\gamma \; \beta \; D \; \frac{f(\chi)}{\frac{1}{4\pi}\int W(\theta)\,d\Omega} \end{aligned} \qquad \text{Eq. (3.1)}$$

Here χ is the angle betweem counters and $f(\chi)$ is the angular correlation, $W(\theta)$, as observed with macroscopic alpha and gamma counters having sizes defined

* For a general discussion of coincidence techniques see 3.4.

by g_α and g_γ. $f(\chi)$ is computed from $W(\theta)$ in Appendix 1. The difference between $f(\chi)$ and $W(\chi)$ is about fifteen percent and can be neglected for qualitative statements about the angular correlation. The integral in the denominator of the expression for C is a normalization over all solid angle.

The formula for R assumes that for all χ,
$\dfrac{f(\chi)\eta_\alpha g_\alpha}{\frac{1}{4\pi}\int W(\theta)\,d\Omega}$ and $\eta_\gamma\, g_\gamma\, \beta\, \dfrac{f(\chi)}{\frac{1}{4\pi}\int W(\theta)\,d\Omega}$ are small compared to unity. These conditions are equivalent to saying that the coincidence rate is a negligible fraction of either individual rate and are satisfied in this experiment. An example of when they would not be met is the counting of coincidences between annhilation radiation at 180° for counters with high intrinsic efficiencies.

The true coincidence rate, C, is obtained experimentally as the difference of the observed coincidence rate (R+C) and R which is computed from the resolving time and individual rates. The square of the standard deviation of C in an observation time, t, is therefore

$$(\Delta C)^2 = \frac{R + C}{t} + \left(\frac{\Delta\tau}{\tau}\right)^2 R^2 \qquad \text{Eq. (3.2)}$$

and the square of the relative error in C will be measured by $\left(\frac{\Delta C}{C}\right)^2 = \frac{1 + R/C}{Ct} + \left(\frac{\Delta\tau}{\tau}\right)^2 \left(\frac{R}{C}\right)^2$

$\frac{R}{C}$ is found from Eq. 3.1 to be $\frac{2\tau D}{q(\chi)}$

where $q(\chi) = \frac{f(\chi)}{\frac{1}{4\pi}\int W(\theta)\, d\Omega}$ and would be unity for an isotropic angular correlation.

$\frac{\Delta\tau}{\tau}$ measures the uncertainty in τ which quantity is assumed to be subject to statistical fluctuations.

Further use of Eq. 3.1 gives,

$$\left(\frac{\Delta C}{C}\right)^2 = \frac{1}{\eta_\alpha g_\alpha \eta_\gamma g_\alpha \beta D q(\chi) \cdot t} + \frac{2\tau D}{q(\chi)} \frac{1}{\eta_\alpha g_\alpha \eta_\gamma g_\gamma \beta D q(\chi) \cdot t} + \left(\frac{2\tau D}{q}\right)^2 \left(\frac{\Delta\tau}{\tau}\right)^2 \qquad \text{Eq. (3.3)}$$

The correlation experiment will not be possible unless the parameters in formula 3.3 be chosen so as to make $\frac{\Delta C}{C}$ acceptably small. The nature of the decay scheme fixes β and determines $q(\chi)$ except for a small dependence on g_α and g_γ. There is always a limit to the observation time, t, if only the patience of the observer. The angular resolution can be sacrificed to build up g_α and g_γ in the familiar conflict of intensity and resolution but there is clearly an early limit in improving accuracy this way. It is most important, particularly for small β to use counters with high intrinsic efficiency.

Resolving time and source strength are important variables. The first term of Eq. 3.3 gives the error for perfect resolution in time of coincidence events and is just the reciprocal of the number of true coincidences counted. The third term recognizes the fact that τ can never be perfectly known and is responsible for the existence of a minimum in the plot of error against source strength. It is clearly desirable to reduce τ until it becomes about the mean life for gamma emission. However the lower limit on τ is usually set by some other consideration as for example the amplifier available or the decay time of the phosphor. The role of source strength may be seen from Fig. 3.2 which is a plot of $\frac{\Delta C}{C}$ against $2\tau D$ for a two hour observation time and for the values of parameters used in the radiothorium correlation experiment. The abscissa is the ratio of random to true coincidences for an isotropic correlation. The value of D to use is not critical so long as it is somewhere near the minimum of the error curve. In general, it is safer to stay somewhat below the value of D which gives the minimum error because it is difficult to be sure that the uncertainty in τ is of a statistical nature.

The error curve shows the radiothorium correlation experiment to be possible, at least fromthis formal stand-

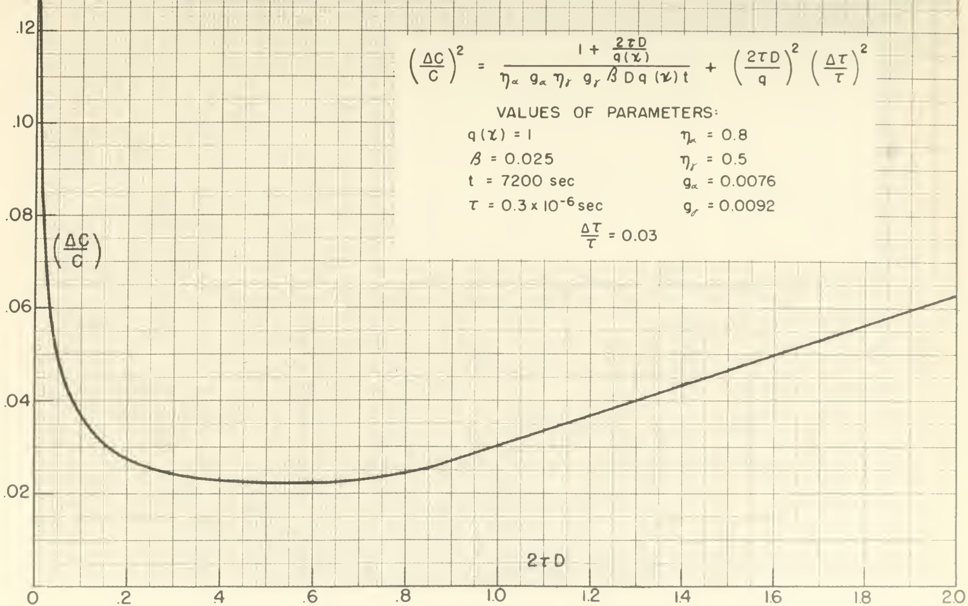


Figure 3.2

THE RELATIVE ERROR OF AN IDEAL COINCIDENCE EXPERIMENT AS A FUNCTION OF SOURCE STRENGTH, D.

point which neglects the existence of the decay products. Two hours counting defines one point to better than three percent. A ten hour run at five angles would therefore give a good idea of the angular correlation if there were no other factors to consider.

Section 2.3. The Alpha and Gamma Detectors

The principal characteristics necessary to the alpha counter, aside from good intrinsic efficiency, were rapidity and reproducibility in the time from the nuclear disintegration until the output pulse reached a value specified by the discriminator setting of the coincidence circuit. A long rise time and particularly a variable one would require an increased resolving time with attendant loss of accuracy. An important secondary requirement was that the alpha counter be relatively insensitive to electrons and gamma rays. The last specification is necessary to prevent counting of beta-gamma, beta-x ray, gamma-gamma and similar coincidences due to the decay products of radiothorium.

A proportional counter with a gas multiplication of the order of a hundred might have been used. Such a detector would not count betas or gammas and, with extremely careful design, a rise time for the electron pulse of a few tenths of a microsecond subject to a jitter of

perhaps a tenth microsecond could be obtained (2.5). This solution was rejected in favor of a scintillation counter because the latter is much easier to make than a proportional counter with the required rise time properties.

The alpha phosphor used was a piece of anthracene about seven mils thick. The crystal was not perpendicular to the alpha particle trajectory so the effective thickness approximated ten mils which exceeded the range of the most energetic alpha particles in the decay chain by more than a factor of two. Alpha particles produce scintillations in anthracene with the pulse size approximately proportional to the alpha energy. The decay time of anthracene is about 3×10^{-8} seconds which is small compared to the rise time of the amplifiers available for this experiment.

Unfortunately, an alpha particle produces only about one eighth as much light in anthracene as an electron of the same energy. The RdTh alpha rays were therefore equivalent to an energy loss in the crystal by betas of 650 kev. The decay products emit beta rays of energy up to 2.25 mev. However, a one mev electron will lose only about 40 kev in a ten mill path length in anthracene. One of low energy and consequent high specific ionization could dissipate about 100 kev to the crystal in going 10 mils.

It is quite easy to discriminate against pulses of this size and still count 650 kev electrons with high efficiency. Despite the foregoing there remains, theoretically, a mechanism for electron counting in the alpha counter. This is scattering of rather low energy electrons resulting in a few electrons, having a path several times the crystal thickness. The counting of electrons in the alpha counter even with an efficiency small compared to that of alphas is potentially serious because the number of gamma and x-rays in coincidence with the electrons produced by decay products is, on the average, much higher than the number of 85 kev gammas per radiothorium alpha. The effect could have been materially reduced by thinning the crystal to three mils. This would have required making a major project of crystal preparation and was not attempted because the seven mil crystal proved good enough.

The requirements for the gamma counter beside good efficiency and rise time were that it should not count particles and that its efficiency as a function of gamma ray energy should have a maximum near 85 kev. The exclusion of beta rays was accomplished by shielding the counter by a quarter of an inch of aluminum. This reduced the intensity of the 85 kev gammas by twenty-eight percent.

The scintillator used in the gamma counter was a thallium activated crystal of sodium iodide one-eighth inch thick. This thickness absorbed about 95% of the incident 85 kev photons principally by photoelectric effect. The main absorption process in NaI for gamma rays of more than a few hundred kev is Compton effect rather than photoeffect so that the absorption coefficient and gamma counter efficiency are much lower for energetic gammas than for the gamma rays from radiothorium. The decay time of a NaI(Tl) phosphor is about half a microsecond. Depending on the amplitude distribution of the scintillations and discriminator setting this sets a lower limit to τ of something like a fifth of a microsecond. The resolving time to be used is decided by weighing the loss of true coincidences in a given reduction of τ against the corresponding beneficial reduction in the random rate. The decision, though important, is not critical and no special effort was made in selecting the value of τ = 0.29 microsecond used in this experiment. The loss of true coincidences due to the long decay time of sodium iodide should be recognized in Eq. 2.1 by multiplying the expression for C by the efficiency of the coincidence circuit. For the angular correlation measurement this efficiency was about seventy percent.

Section 3.4. Arrangement of the Counters

It is important to keep the source fixed relative to the alpha counter as the angle between counters is varied in order to prevent the alpha counter efficiency from changing with angle. The gamma counter must be capable of motion relative to the alpha counter about the source as center. It was decided to keep the gamma counter fixed and move source and alpha detector together. The source was thin and mounted on a one mil polystyrene backing having negligible absorption so that the gamma counter efficiency was independent of angle except close to 90° and 270° where a light aluminum clamp which held the polystyrene interfered.

The decision on the proper angular resolution for a correlation experiment is rather subjective. Here a half angle of ten degrees was used in the alpha counter and eleven degrees in the gamma detector. This made $g_\alpha = 0.0076$ and $g_\gamma = 0.0092$ both of which are surely too small considering that lack of intensity is one of the main problems in the radiothorium experiment.

It is desirable to have the source and counters housed in a relatively large volume so that radioactive contamination which accumulates on the walls will have small solid angle for counting and any other wall effects will be minimized. For this experiment the detectors and source

were in an evacuated cylindrical chamber seven inches in diameter by seven inches high. A uniform contamination on the walls equal to the activity of the source would have given a counting rate of considerably less than one percent of that due to the source.

Scattering of gamma rays is frequently a worry in angular correlation measurements. In the radiothorium experiment, it presented no problem. The entire chamber was lined with a thirty second of an inch of lead which was sufficient to absorb virtually all 85 kev gamma rays striking it. The absorption process in lead is photo-electric effect in the L shell and the L x-rays produced are absorbed, by aluminum, before reaching the gamma counter. Consequently, the only 85 kev gamma rays counted are those emitted into the collimator covering the gamma counter.

Figures 3.3 and 3.4 show the equipment, including arrangement of the source and counters.

Section 3.5. Electronic Equipment

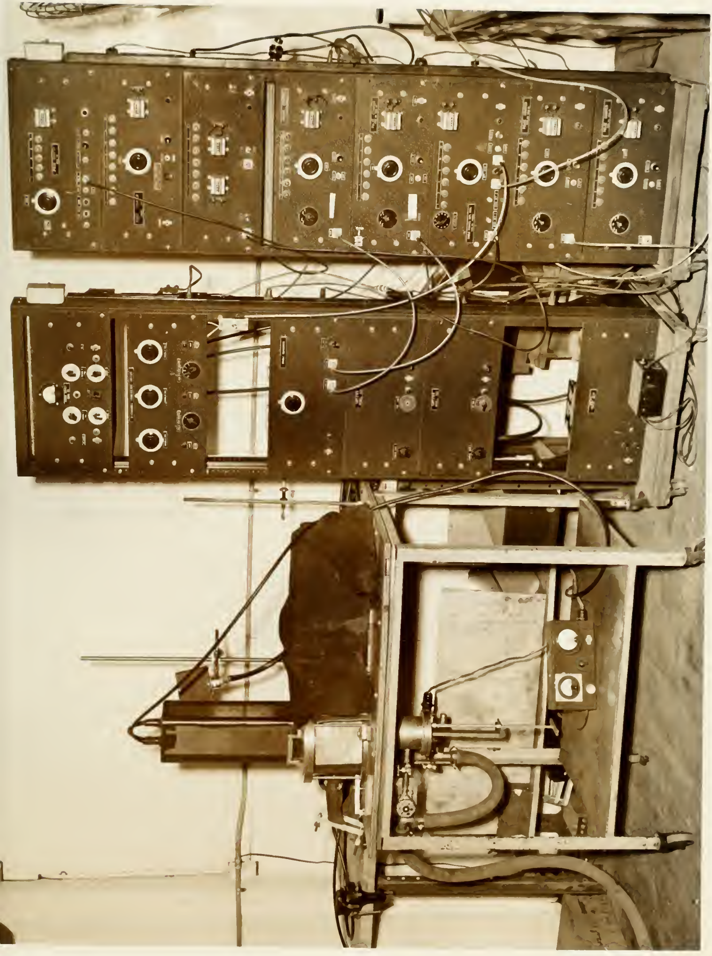
The electronics for the angular correlation measurement was an assembly of standard components most of which are described in 3.6. Figure 3.5 shows their interconnections schematically. The amplifiers had 0 to 100% rise times of 0.18 x 10^{-6} sec. and were delay line clipped

FIGURE 3.3

(a) General Equipment.

(b) Assembled Vacuum Chamber with Counters and Air Lock for the Introduction of Absorbers.

(c) Interior View of Vacuum Chamber Looking Toward the Gamma Counter. Air Lock Door Open.



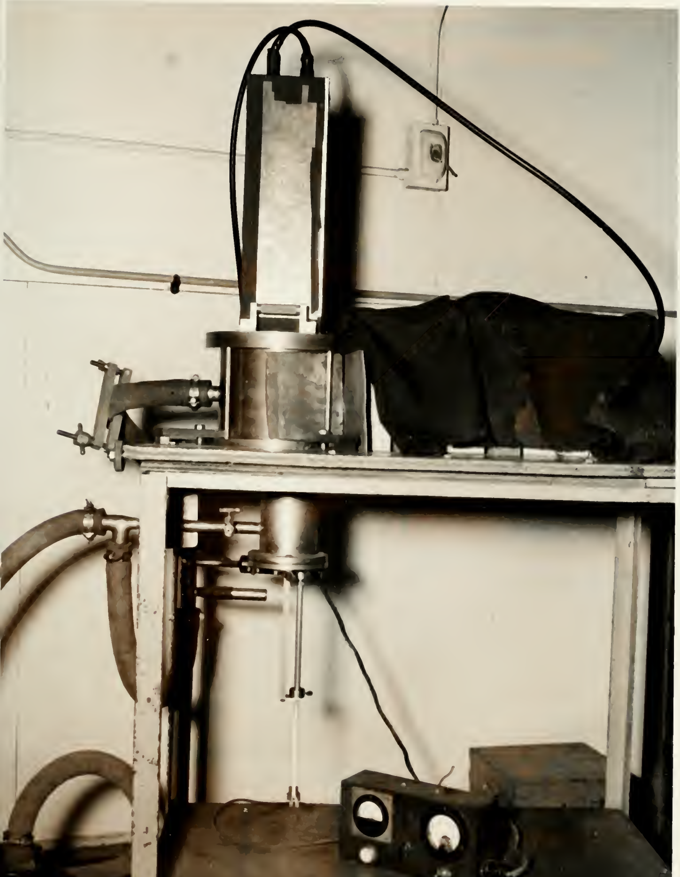
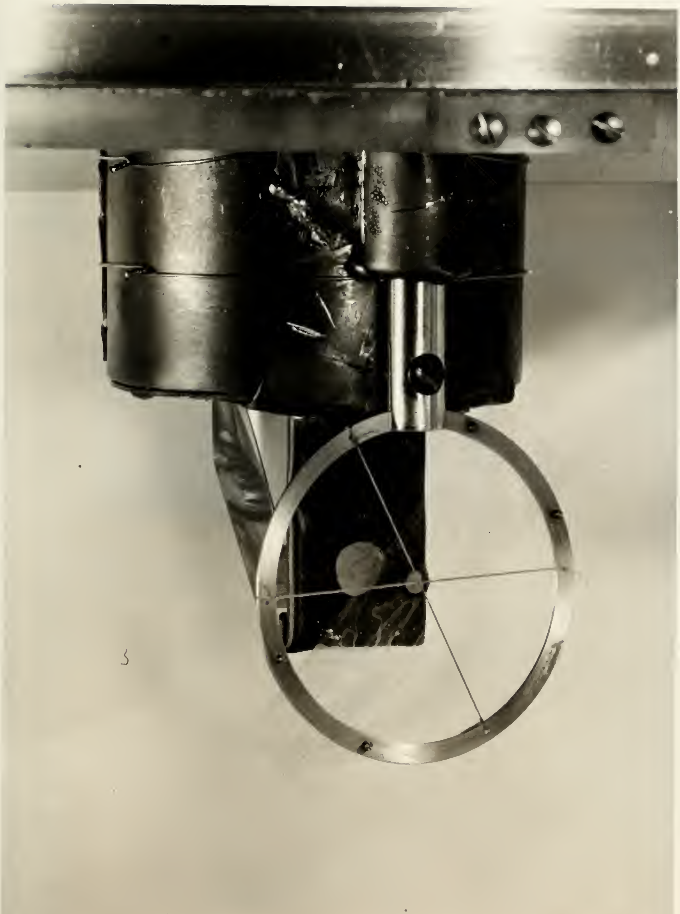


Figure 3.4

Alpha Light Pipe, Collimator and Source. An electrically operated 2 mil copper shield capable of covering the hole in the alpha collimator has been omitted from the figure.



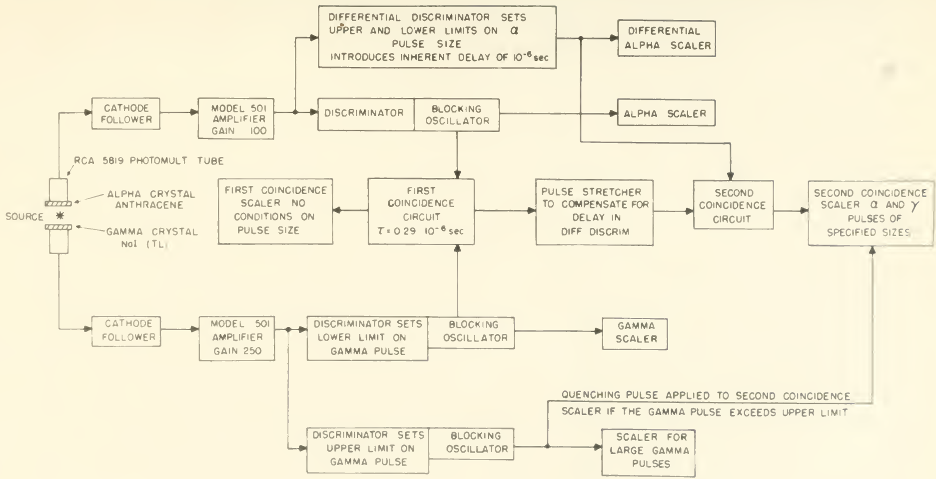


Figure 3.5

SCHEMATIC DIAGRAM OF ELECTRONIC EQUIPMENT

to produce square pulses a microsecond wide for use with a differential discriminator. This instrument was used to measure the pulse height distributions of the counters and also to set upper and lower limits on the pulses from the alpha counter in the correlation measurement. The amplifiers had crystal diodes, connected after the manner of Cross (3.7), to prevent multiple pulsing when overloaded. Some such limiting feature was essential because of the considerable energetic gamma radiation produced by the decay products.

CHAPTER IV

The Experiment

Section 4.1. Introduction

A measurement of the radiothorium angular correlation was attempted by Kulchitski et alia (4.1). The authors say that their results are only qualitative because the experiment was performed on radiothorium in equilibrium with its derivatives and some of the coincidences were caused by the alpha decay of ThC. Even if their data are accepted as due to radiothorium, the errors are so large that the question of whether or not the correlation has a $\cos^6\theta$ term comparable to the lower powers of $\cos^2\theta$ which are present cannot be settled. The inaccuracy can be traced primarily to the low intrinsic efficiency of the geiger counter used to detect gamma rays. Nevertheless, a much better experiment could have been done had they separated radiothorium from its decay products. The use of a built up source gave a random coincidence rate about ten times the true rate even for a weak source. Had ten separations been made and data taken for the first five hours after separation, the correlation could have been determined to much better than ten percent at each of five angles.

In the present correlation measurement chemical separation was used, and data was taken for ten hours. The experiment soon revealed itself as a battle to keep down the growth of true coincidences from the decay products which began after the separation. These were mainly beta-gamma coincidences with the beta rays counting in the alpha counter. The exclusion of spurious coincidences was eventually accomplished well enough so that a measurement of the radiothorium angular correlation could probably have been made when RdTh was in equilibrium with its decay products.

Section 4.2. The Chemical Separation

Radiothorium was purchased* as chloride free of carrier except for a tenth of a milligram of iron per millicurie. Elements preceding RdTh in the thorium series were absent. For each separation a few drops of the original solution were evaporated to dryness and the residue taken up in 0.25 ml of 0.1N HNO_3. The RdTh solution was then mixed with freshly prepared, well washed, $PbSO_4$ and PbS precipitates and heated to 80°C. for fifteen minutes in the presence of excess H_2SO_4. The $PbSO_4$ was responsible for the precipitation of ThB and ThX which are Pb and Ra isotopes respectively, and the PbS insured precipitation of the Bi isotope ThC. Radiothorium and ThC" remained in

* From Fleischman Burd & Co., 22 West 48th St., New York, 19, N.Y.

the supernatant which was decanted after cooling and centrifuging. The foregoing separation was then repeated, the iron extracted with ether, and the RdTh solution evaporated to dryness on the polystyrene backing. The freshly prepared source of radiothorium chloride was covered with approximately 0.5 mg/cm^2 of shellac as insurance against flaking in the vacuum.

Section 4.3. Tests for Radiothorium

It was, of course, essential to verify that the initial radioactive material was radiothorium. A number of tests were made and most are briefly itemized below.

1. The growth of activity of gamma rays more energetic than one mev followed the theoretical curve for ThC. Figure 4.1 shows this curve which was derived neglecting the effect of the one hour half life of ThC.

2. The growth of coincidences was followed to eight hundred hours and lay on the curve.

3. The residual activity when an unshellacked source was removed after having

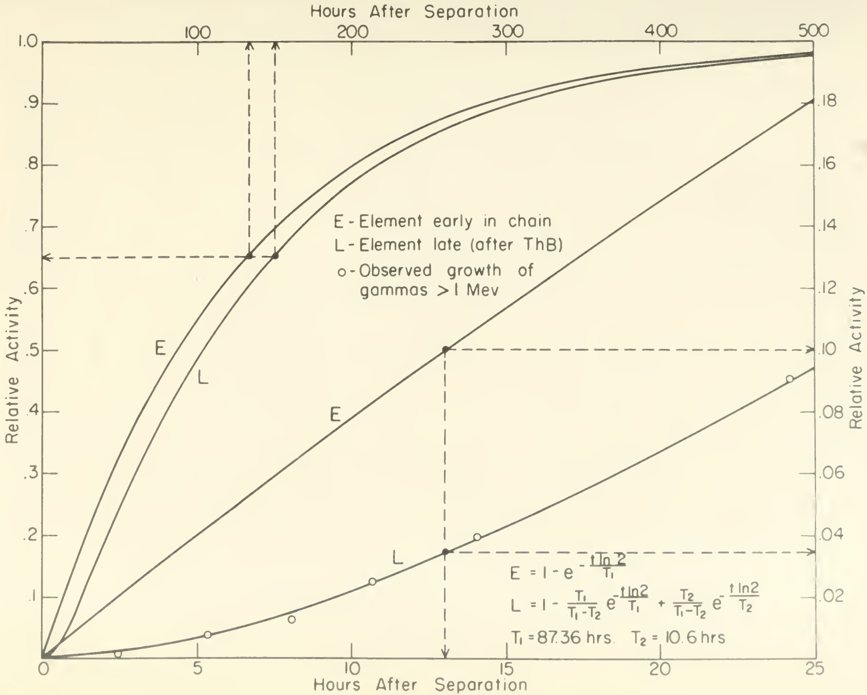


Figure 4.1
GROWTH OF ACTIVITY IN THE RADIOTHORUM DECAY CHAIN

been in the vacuum chamber for a day was followed for three days and disappeared with the 10.6 hour activity of ThB produced by thoron decay.

4. When the PbS was omitted from the chemical treatment so that ThC should not have been separated a strong one hour beta activity was found. Absorption in aluminum showed an end point of 2.2 mev.

5. A three minute gamma activity was observed to die away after a separation. This was in accord with the chemistry which should not have removed ThC".

6. The differential bias curve for the alpha counter, taken at separation, showed a single peak at the position of the peak due to the 5.3 mev alphas from Po^{210}. Several weeks later the original peak had broadened on the high energy side. This was consistent with the fact that the alpha particles from radiothorium are less energetic than those of its decay products. A second, well resolved,

peak occurred in the late time bias curve at the approximate position expected for the 8.78 mev ThC' alphas, and with the correct relative intensity.

The measurements just described leave no doubt that the experiment was performed on radiothorium. It is probable that they were not sensitive enough tests to reveal a small admixture of other radioactive elements. The contaminant most to be feared is one having alpha fine structure such as ionium, for example. However, strong plausibility arguments can be made against the presence of known alpha emitters with fine structure in amounts sufficient to affect the correlation.

During the course of the experiment a number of ampules of radioactive material were purchased as radiothorium. Care was taken to test the contents of each one in more than one way.

Section 4.4. The Suppression of Spurious Coincidences

The first attempt at the correlation measurement was made with crude equipment. All pulses from the counters exceeding specified minimums were fed to the coincidence

circuit after pulse shaping. The anthracene crystal for alpha detection was about twenty mils thick and was poorly attached with lucite solvent to a five inch lucite light pipe which led to the photomultiplier tube. The sodium iodide crystal had not been cleaned of the hydrated NaI surface covering and a 2.5 inch light pipe was used. Both counters had amorphous bias curves. Figure 4.2 shows the coincidence rates at 180° and 136° as functions of time after separation for an early run. No significance should be attached to the apparent slower build-up at 180° because of the large errors. Figure 4.3 gives the angular correlation obtained from the first six points of the run. Each point represents forty minutes counting and the average time after separation was six hours. The curve through the points of Fig. 4.3 is the least squares fit obtained from four runs made much later with elaborated equipment. It represents the final measurement of the combined angular correlation of both gamma rays. Figures 4.2 and 4.3 show that the early equipment was on the borderline of being good enough for a measurement of the ordinary correlation. However, it was not good enough for separation of the individual correlations by the thallium absorption technique. This required reduction of the intensity of the gamma rays from the radiothorium decay by a factor of three without

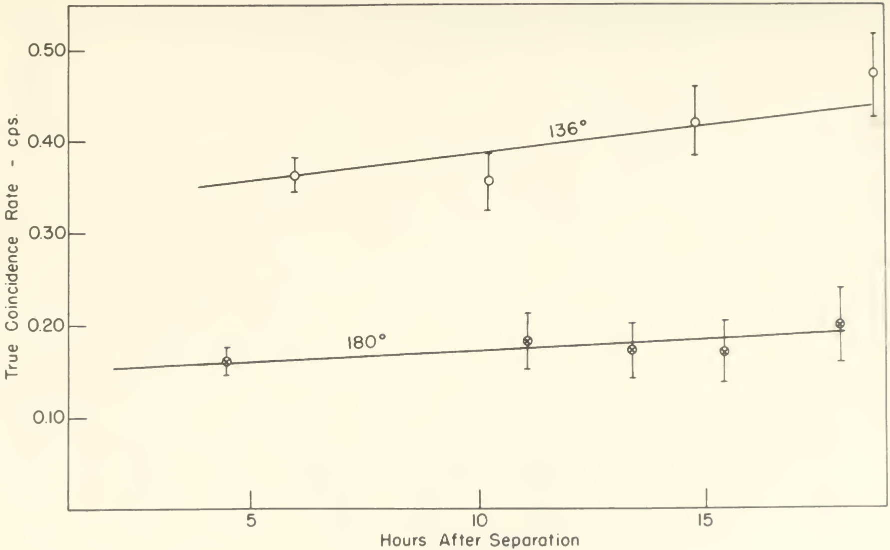


Figure 4.2
TRUE COINCIDENCE RATES PLOTTED AGAINST SOURCE AGE FOR AN EARLY CORRELATION RUN.

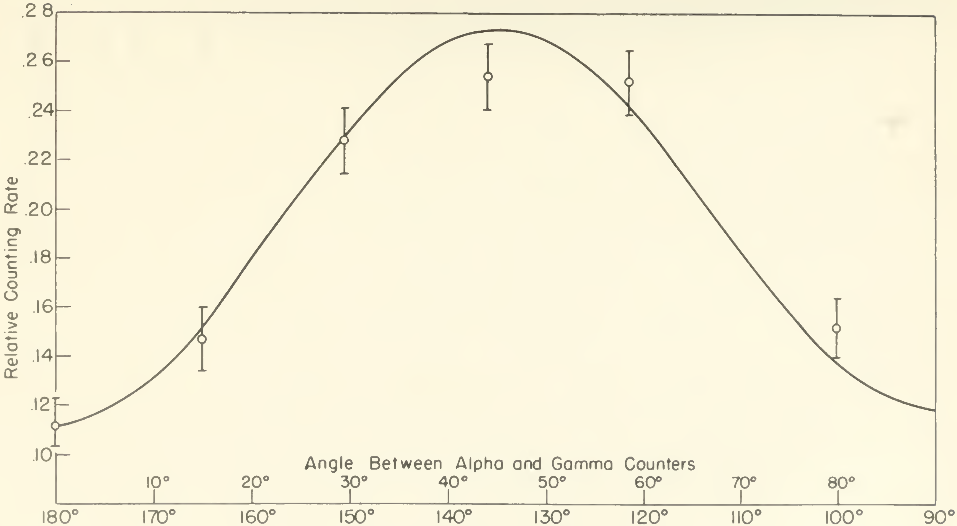


Figure 4.3

AN EARLY ANGULAR CORRELATION RUN. 40 MINUTES COUNTING PER POINT. THE CURVE IS THE LEAST SQUARES FIT TO THE FINAL DATA OF THE EXPERIMENT

a corresponding cut in other gammas and made build up much more of a problem.

The first step in improving the instrumentation was the exclusion of coincidences between the 8.78 mev alpha particles from ThC' and gamma rays from excited states of ThC'. The coincidences could occur because of the 3×10^{-7} sec. half life of ThC' and were removed by refusing to count coincidences due to energetic alpha particles. A differential discriminator was incorporated in the alpha channel as indicated in Fig. 3.5. The alpha counter was redesigned to sharpen the bias curve with the light pipe shortened to about 1.5 in., the crystal thinned to seven mils and its coupling to the light pipe much improved. The degree of alpha energy resolution with the new equipment applied to a rather thick source is shown in Fig. 4.4. Figure 4.5 is a bias curve of the same source three days later when the decay products were half grown. The photomultiplier voltage was reduced for this run to get the ThC' alpha peak and the main peak on the discriminator dial range.

The effect of the modification was two-fold. It reduced the build up due to betas in the alpha counter, primarily because of the thinner crystal, and it eliminated ThC as a source of trouble because of the improved bias curve. The overall result was to diminish the growth of

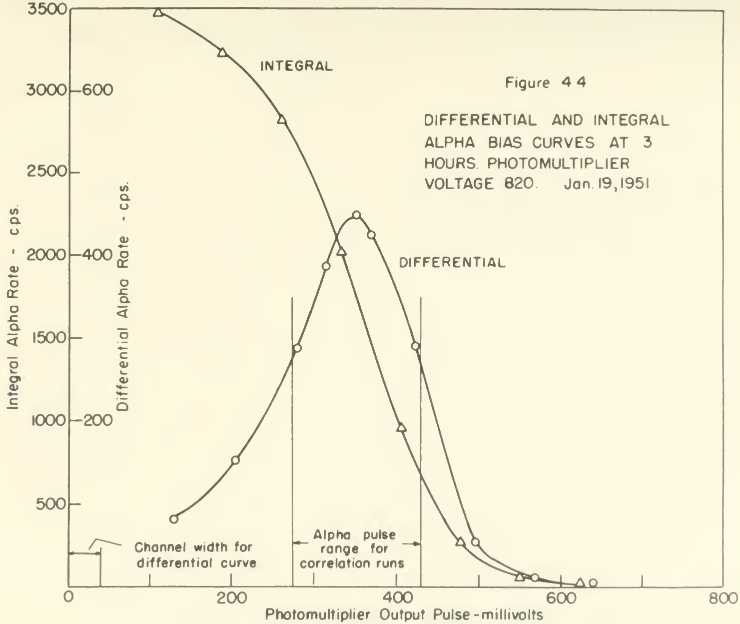


Figure 4 4

DIFFERENTIAL AND INTEGRAL ALPHA BIAS CURVES AT 3 HOURS. PHOTOMULTIPLIER VOLTAGE 820. Jan. 19, 1951

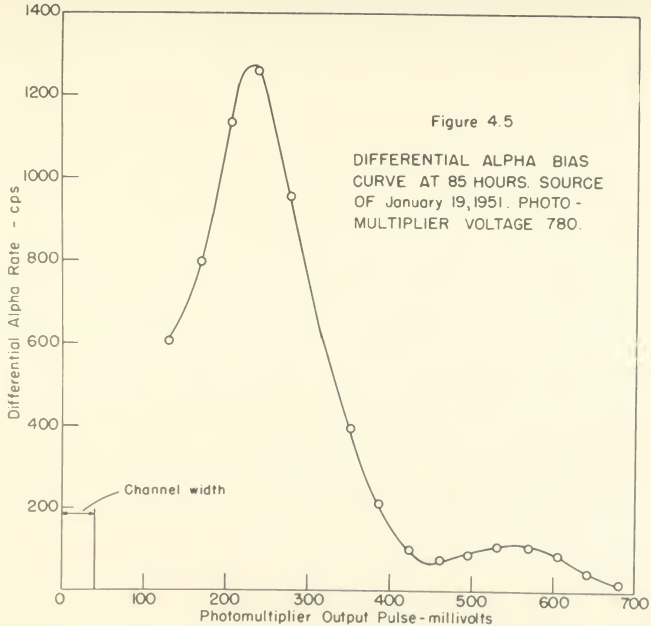


Figure 4.5

DIFFERENTIAL ALPHA BIAS CURVE AT 85 HOURS. SOURCE OF January 19, 1951. PHOTO-MULTIPLIER VOLTAGE 780.

true coincidences to about a third the original value without loss in the true coincidence rate from radiothorium.

There still remained a small increase in the coincidence rate in the first ten hours. It was eliminated, to an extent stated in Section 4.5, by introducing an upper limit on gamma pulse size. This was profitable because, after cleaning the sodium iodide crystal and shortening the gamma light pipe to a little over an inch, fairly good bias curves were obtained. One is illustrated in Fig. 4.6 for a radiothorium source three hours after separation. The counting of coincidences only when the gamma pulse was less than a specified size was accomplished by applying a quenching pulse to the inverter of the coincidence scaler whenever a gamma counter pulse was big enough to trigger a discriminator set at the critical voltage.

The condition on alpha pulse height completely removed coincidences from ThC' but was of no help with ThC and ThX whose alpha particles could not be resolved from those of radiothorium with this apparatus. The 40 kev gamma ray from the ThC fine structure was eliminated, at a sacrifice of twenty percent of the 85 kev gamma rays, by placing 4.5 mils of tin in front of the gamma collimator. ThX was left as the only possible source of alpha gamma coincidences in the daughters.

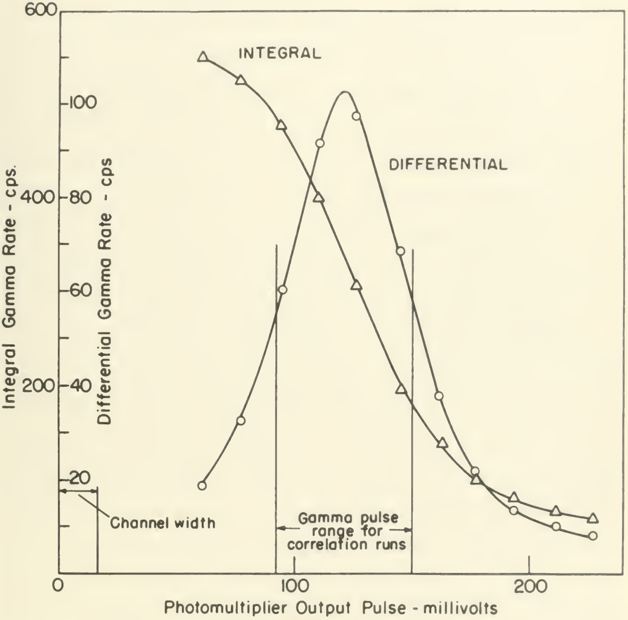


Figure 4.6

DIFFERENTIAL AND INTEGRAL GAMMA BIAS CURVES AT 3 HOURS. LARGE SOLID ANGLE. March 23, 1951.

Section 4.5. Correction for Coincidences from Decay Products

The gamma ray of about 250 kev to be expected from the alpha fine structure of ThX has not yet been observed although its conversion electrons have been sought (4.2). Presuming it were unconverted, it would have five percent intensity in equilibrium compared with .025 for the 85 kev γs. Assume these had intrinsic detection efficiency of only one half, because of the aluminum and tin absorbers, which did not affect the 250 kev gamma. About ten percent of the 250 kev photons would have had a Compton interaction in the one-eighth inch NaI crystal and those absorbed by photoeffect should be ignored because they would have made pulses greater than the maximum size allowed to count. The gate width in the gamma channel was about 50 kev and the differential Compton cross section is roughly independent of energy below 125 kev. The maximum counting rate at equilibrium for gamma rays from the ThX decay relative to that of the 85 kev quanta, therefore, can be estimated as $2(\frac{.05}{.025}\cdot\frac{1}{10}\cdot\frac{50}{125}) = \frac{1}{6}$. From Fig. 4.1 the relative intensity at ten hours would have been $0.08\cdot\frac{1}{6}$ or about a percent. The effect would have been three times as big in an absorption run but still not very important.

The growth of coincidences for a weak source was followed essentially to completion and the angular correlation was measured with the decay chain near equilibrium. See Figs. 4.7 and 4.8. The early time correlation appeared superimposed on an isotropic correlation of about two and a half times the intensity. Only at most a fifteenth of this growth could be ascribed to ThX by the argument above. The build up of spurious coincidences was, therefore, mainly due to electrons counting in the alpha counter and could be corrected for by curve L, Fig. 4.1. The correction for a ten hour observation is a subtraction of an isotropic term of 1.8% for ordinary runs and 5.5% for runs with absorber. It was ignored except for the absorbed runs.

Section 4.6. Work with the Gamma Rays

The evidence that the 83.3 and 86.8 kev gamma rays are not emitted in cascade has previously come entirely from alpha fine structure. It was decided to look for coincidences between 85 kev gamma rays to be sure the decay was not in two steps. This was done with separate counters having solid angles about twenty times larger than the angular correlation counters but otherwise identical to the usual gamma detector. Upper and lower limits were set on pulse size, and the source was counted for normali-

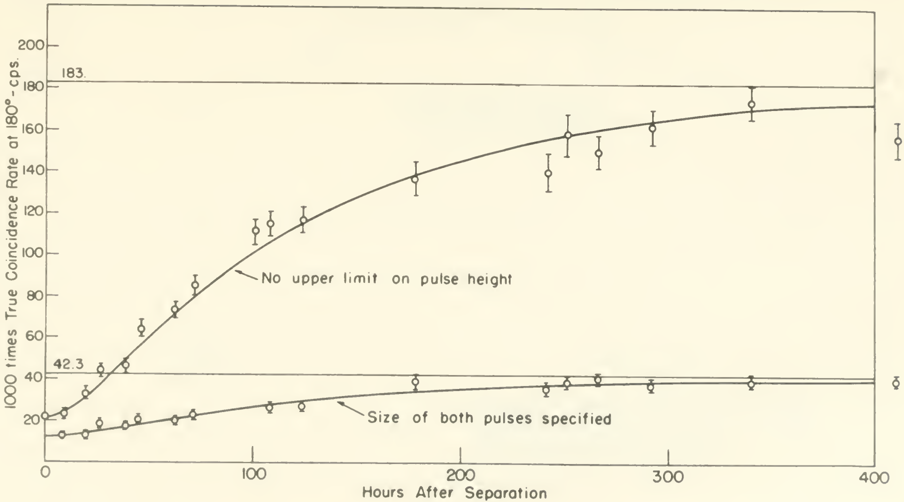


Figure 4.7

TRUE COINCIDENCE RATE AGAINST TIME FOR A WEAK SOURCE SEPARATED February 7, 1951. CURVES SHOW THEORETICAL GROWTH OF ThB AND ThC.

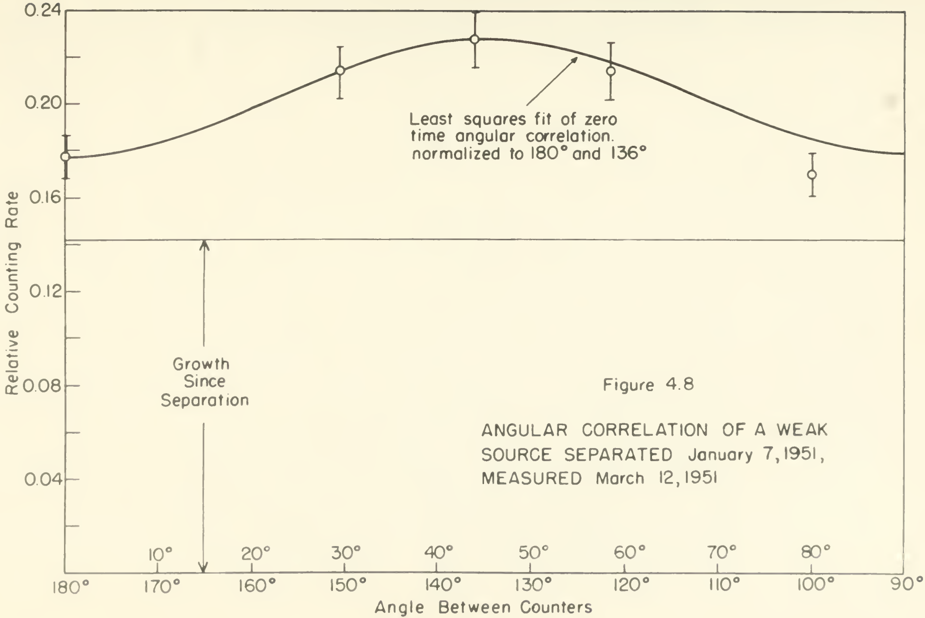


Figure 4.8

ANGULAR CORRELATION OF A WEAK SOURCE SEPARATED January 7, 1951, MEASURED March 12, 1951

zation in the alpha-gamma apparatus. The gamma-gamma coincidence rate was one percent of the alpha-gamma rate and can probably be attributed to a not quite perfect chemical separation.

The average energy of the gamma rays in coincidence with alpha particles was confirmed by absorption in lead and gold as lying between the K absorption edges of those elements. Because of the small geometrical efficiencies of the counters, this fixing of the energy as between 81 and 88.5 kev is only accurate to about five percent. The correlation measurements with thallium absorber showed a change from the correlation without absorber. Since runs with lead and gold absorbers gave the same correlation as runs without absorber,* the alteration by thallium is evidence of a component between the K edges of lead and thallium. This component must be of low intensity since the measured absorption coefficients in lead and thallium were the same, within error, and about a third of the gold absorption coefficient. This experiment, therefore, gave no reason to doubt the gamma ray energies and relative intensities of Surugue and Tsien and Riou.

Section 4.7. Angular Correlation Runs

The essential equipment for the measurement of an alpha-gamma angular correlation is simple so it is a

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* See Fig. 4.14, next page.

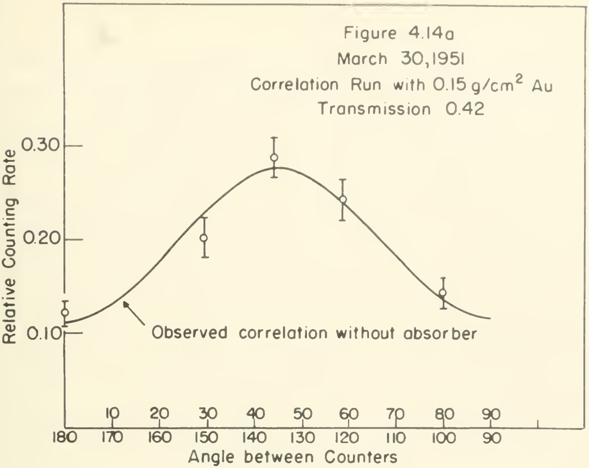
Figure 4.14a
March 30,1951
Correlation Run with 0.15 g/cm² Au
Transmission 0.42
Relative Counting Rate
0.30
0.20
0.10
Observed correlation without absorber
10 20 30 40 50 60 70 80 90
180 170 160 150 140 130 120 110 100 90
Angle between Counters

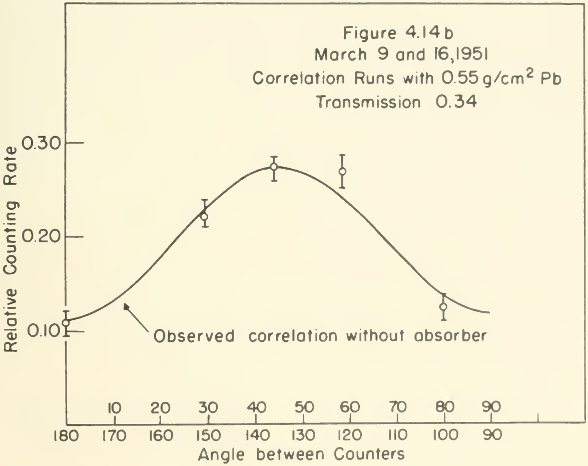
Figure 4.14 b
March 9 and 16,1951
Correlation Runs with 0.55 g/cm² Pb
Transmission 0.34
Relative Counting Rate
0.30
0.20
0.10
Observed correlation without absorber
10 20 30 40 50 60 70 80 90
180 170 160 150 140 130 120 110 100 90
Angle between Counters

temptation to assume that there will be little systematic error which depends on angular position. Nevertheless, the apparatus must be checked to allow any confidence in the result of the experiment. The most desirable test would be the measurement of an angular correlation which had been confirmed by a number of observers. Outside of nuclear reactions which yield alpha particles*, there are as yet no firmly established angular correlations between alpha and gamma rays. There has been a recent measurement of the correlation between alpha particles and 40 kev gamma rays in the ThC $\rightarrow$ ThC" transition (4.6). Even if it is assumed that this experiment is reliable within the errors quoted, it is not suitable for calibration except as a check to the same accuracy. This is because interpretation of the measured correlation will probably require that the alpha particles be emitted as a mixture of different orbital angular momenta making the correlation dependent on the unknown matrix elements involved in alpha emission.

A reasonably direct test of the equipment was made on some of the known gamma-gamma correlations. The thin alpha crystal was replaced by a thicker piece of anthracene for this purpose and the correlation of Na^{22} annihilation quanta was observed. The result confirmed the designed

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* for alpha-gamma correlation experiments in reactions, see 4.3, 4.4, 4.5.

angular resolution of the counters. The gamma-gamma correlation in Pd^{106} was also measured and agreed with the results of Brady and Deutsch (4.7).

It is important to take data in such a way that it will provide a direct check on equipment and so that systematic errors of one sort or another will tend to average to zero. Since $W(\theta)$ is the sum of powers of $\cos^2\theta$ the observed correlation should be symmetrical about 0°-180° and 90°-270° axes. The symmetry is an excellent test of the equipment and it is desirable to make a complete check of it for each ten hour run. About twenty minutes were needed to get significant data and each basic angle had, in general, four reflections so that not more than about seven angles could be thoroughly checked for symmetry in a run. This number is more than enough to define any correlation which can be resolved experimentally and five angles were usually picked. The observed correlation was symmetric to the accuracy of the data which was three percent for runs without absorber.

Because of the growth of the alpha and gamma counter rates with time there was no simple way of normalizing the coincidence rate to the product of that part of the individual rates which was due to radiothorium. Such a normalization is desirable because it corrects for

changes in alpha and gamma counter efficiences. It is not essential because efficiency changes are small compared to statistical errors and should average out in a large number of observations carried out over a considerable period of time. Tests with Po^{210} and Co^{57} showed that the individual rates were isotropic so there was no systematic variation of efficiency with angle. This was confirmed by plotting the alpha and gamma rates as functions of time during the runs.

Resolving time was measured with the radiothorium source by introducing a two microsecond delay in the gamma channel, and also alternatively by feeding the output of a pulserto one line while the other counted alpha or gamma rays. The two methods agreed completely. However, a possible source of error in a correlation experiment is a large systematic error in the random rate which is isotropic. This was eliminated as a worry because the angular correlation was unaffected by variation of source strength by a factor of five.

The fact that the radiothorium was carried on iron suggests the possibility that if the iron were not extracted perfectly when preparing the source there might be a magnetic interaction tending to reorient the magnetic moment of the excited state of ThX before gamma emission and alter the correlation. To test for this, the correlation

without absorber was measured on a source from which the iron was extracted only very poorly and on a source which had never contained iron, thorium being the carrier. For both cases $W(\theta)$ was the function normally observed without absorber.

Section 4.8. Treatment of Data

For the runs without absorber, coincidences were counted at five basic angles equally spaced in $\cos^2\chi$ and lying between 100° and 180°. Since the same angles were used for each run, point by point combination of runs was possible. The true coincidence rate for a given angle included with its reflections was normalized by dividing by the sum of the five coincidence rates for the run. The probable error asserted was the standard deviation of the true rate computed from equation 3.2 and slightly increased by the propagation of error in the normalization. The final relative counting rate at a given angle was the weighted average of the rates at that angle for the four individual runs. The observed angular correlation was the unweighted least squares fit of this data to the function $a + b \cos^2\chi + c \cos^4\chi$. The fit represented the character of the data showing the correlation did not contain terms of higher power. The transformation between θ and χ, derived in Appendix 1,

was applied in the inverse direction to give the $W(\theta)$ corresponding to the observed correlation. Corrections for Doppler effect, transformation from laboratory to center of mass system and source size were omitted as unimportant.

Two of the runs with thallium absorber were made at five angles, and the other two at seven angles. For each group $W(\theta)$ was found by the procedure just described and a simple average taken after normalization.

Section 4.9. Results

In this section a number of plots of correlation functions against angle are presented. These functions are of two types distinguished by the rather imprecise terms, observed and experimental correlations. An observed correlation is intended to mean a quantity proportional to the true radiothorium coincidence rate observed with the finite counters. It is essentially the least squares fit to the data and is a function of the angle between counters, χ. An experimental correlation is an observed correlation which has been corrected for counter size and is the best estimate of $W(\theta)$ which the experiment allows. Observed correlations are usually shown normalized to the sum of the true coincidence rates for the five angles of observation, while experi-

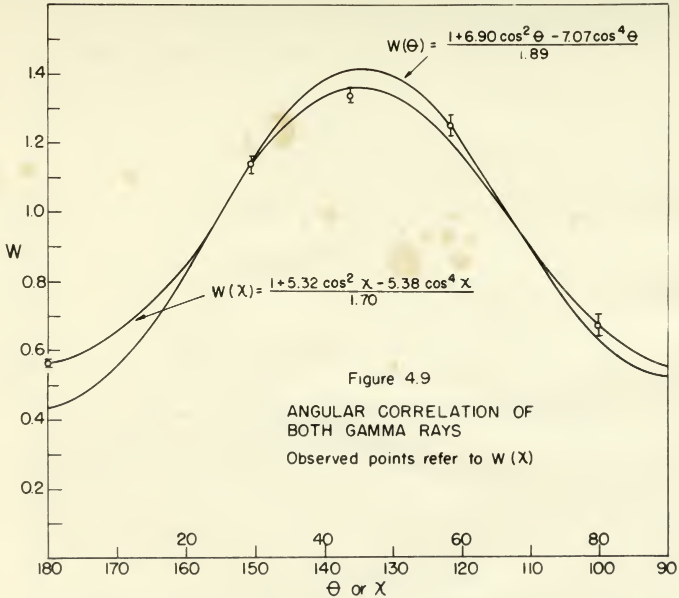


Figure 4.9

ANGULAR CORRELATION OF BOTH GAMMA RAYS

Observed points refer to W (χ)

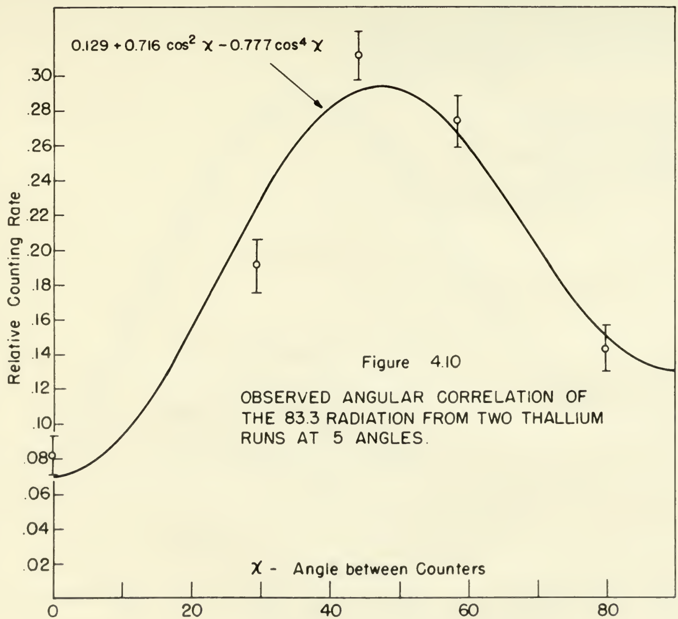


Figure 4.10

OBSERVED ANGULAR CORRELATION OF THE 83.3 RADIATION FROM TWO THALLIUM RUNS AT 5 ANGLES.

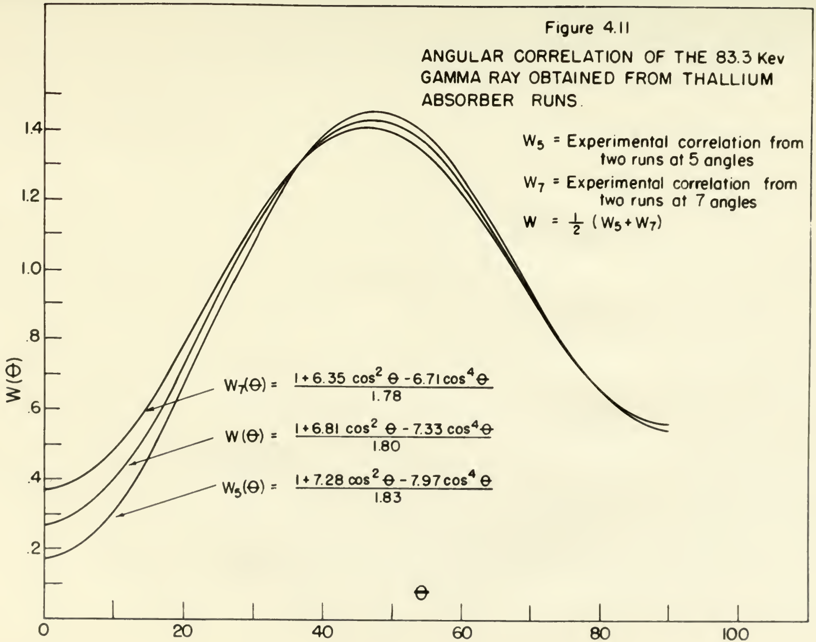


Figure 4.11

ANGULAR CORRELATION OF THE 83.3 Kev GAMMA RAY OBTAINED FROM THALLIUM ABSORBER RUNS.

ray. The five and seven angle runs agree well except near $\theta = 0$ where virtually the whole disagreement occurs. At this angle the individual curves differ from the average by less than two probable errors so there is no definite indication of a systematic error but there is certainly the suggestion of one.

The correlation for the 86.8 kev radiation is given in Fig. 4.12. It was obtained by finding the function which, when added to $W(\theta)$ for the 83.3 kev gamma ray with proper normalization and Riou's estimate of the relative intensity as 0.026/0.074, gave the correlation measured without absorber. The figure shows the effect on this correlation of using the five and seven angle thallium data separately. The validity of the subtraction process used to get the correlation of the 86.8 kev radiation may be questioned. Beside the assumption that Riou is right about the relative intensities, it is necessary to assume that the lifetime of the 86.8 kev gamma is such that it has the same probability of making a coincidence with the preceding alpha particle as the 83.3 kev gamma. If the accuracy of the thallium data were a factor of two better, the relative intensity of the gammas as observed in coincidence with alpha particles and with the resolving time used in the experiment could

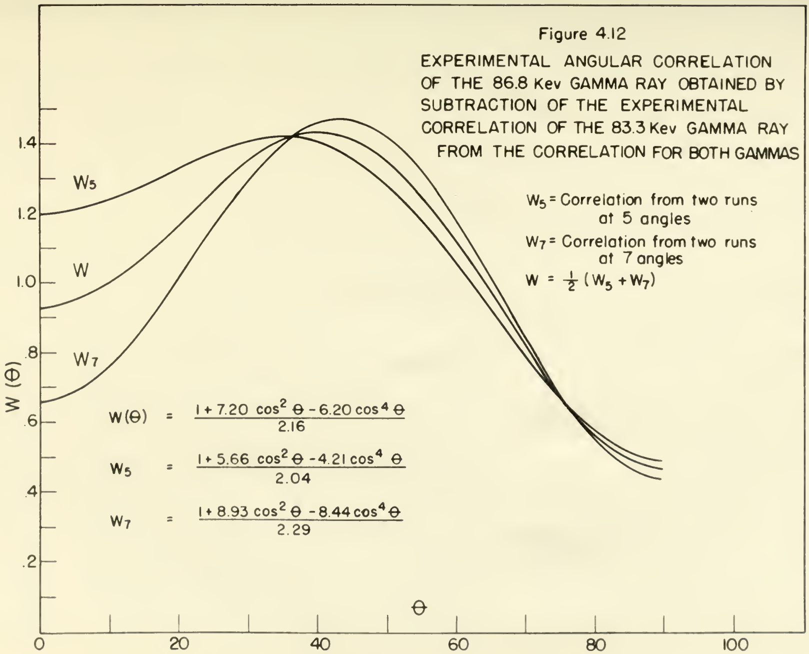


Figure 4.12

EXPERIMENTAL ANGULAR CORRELATION OF THE 86.8 Kev GAMMA RAY OBTAINED BY SUBTRACTION OF THE EXPERIMENTAL CORRELATION OF THE 83.3 Kev GAMMA RAY FROM THE CORRELATION FOR BOTH GAMMAS

be computed from the observed absorption coefficient for coincidences. In support of the subtraction procedure used, it may be argued that since combination of Surugue and Tsien's number of electrons per disintegration for each gamma ray with Riou's numbers of quanta gives nearly identical conversion coefficients for the 82.3 and 86.8 kev radiations these must be of essentially the same multipole nature. They would therefore be expected to have similar lifetimes and be treated symmetrically by the coincidence circuit.

It is desirable to give a single figure as a rough indication of the accuracy of the experimental correlations. The statistical error may be approximated as the percent error of an individual point divided by the square root of the number of angles at which measurements were taken. For the over all correlation, this is about $\frac{4}{\sqrt{5}} = 2\%$. The error of the thallium correlation would be $\frac{10}{\sqrt{5}} = 5\%$ and will be arbitrarily stated as 8% because of the poor reproducibility near zero degrees. It is more difficult to put a limit on the error of the correlation of the 86.8 kev gamma ray. If the assumptions involved in the subtraction by which it is obtained are justified, it is probably correct to thirty percent.

Section 4.10 summarizes the experimental correlations in analytic form. This raises the question of the accuracy of the coefficients in the expansion of the correlations

as powers of $\cos^2\theta$. The question is rather academic because the reason for asking it is to estimate the accuracy of the experimental correlations. This is best done from a consideration of the probable errors of the experimental points and the reproducibility of runs in the manner discussed above. An estimate on the basis of the probable errors of the coefficients is, of course, possible but the natural tendency in making one is to neglect the fact that errors of different coefficients are related and so exaggerate the error. After normalization to unity at 90° the probable errors of the $\cos^2\theta$ and $\cos^4\theta$ terms in the experimental angular correlation of the 87.7 kev radiation are about ten percent. The error in the coefficients of the correlation for both gamma rays is about three percent. In either function the $\cos^4\theta$ term is definitely greater in magnitude than the $\cos^2\theta$ term.

Section 4.10. Summary

The experimental results and some immediate conclusions are summarized below. The next chapter is concerned with conclusions which need discussion.

Results:

(1) Alpha-gamma coincidences were observed with a resolving time of 0.3×10^{-6} sec. The number of coincidences per alpha ray was

consistent with Riou's measurement of one quantum emitted in forty disintegrations of radiothorium.

(2) The gamma ray energy was fixed to an accuracy of five percent as lying between the K absorption edges of gold and lead (81-88.5 kev).

(3) The existence of a gamma ray of low relative intensity and with energy between the K edges of thallium and lead (86.1-88.5 kev.) was inferred from the observed alteration of the angular correlation when a thallium absorber was placed over the gamma counter.

(4) Results (1)-(3) are in agreement with previous experiments where gamma rays of 83.3 and 86.8 kev with relative intensities of 0.74 and 0.26 have been detected. Results (1) and (3) constitute a rough lifetime measurement and set an upper limit to the emission time of both gamma rays of something like 0.5×10^{-6} sec.

(5) Coincidences between gamma rays could not be observed; consequently, the gamma rays are not emitted in cascade.

(6) The angular correlation of both gamma rays, taken together, was measured to two percent as $1 + 6.90 \cos^2\theta - 7.07 \cos^4\theta$.

(7) The angular correlation of the 82.3 kev quanta was found to be $1+6.81\cos^2\theta-7.22\cos^4\theta$ and has a probable error of eight percent.

(8) The angular correlation of the 86.8 kev radiation was obtained by subtraction after normalization as $1 + 7.2 \cos^2\theta - 6.2 \cos^4\theta$. If the assumptions made in the subtraction are justified it is probably correct to thirty percent.

CHAPTER V

Discussion of Results

Section 5.1. Introduction

Angular correlation measurements are called upon mainly for spin assignments, although they may also be capable of giving magnetic moments of excited nuclear states (5.1). For fruitful application of the correlation technique, the sequence and energies of the radiations in the decay scheme should be known. When the radiations emitted in a cascade are not mixed and reorientation is not a problem, the angular correlation depends only on kinematics and can be calculated from well established principles of quantum mechanics. If it is impossible to find an assignment of nuclear spins and angular momenta for the particles emitted in cascade which gives a theoretical correlation agreeing with the experiment the latter must be in error, or the decay scheme must not, in fact, be the simple one assumed in computing the correlations. The relative uniqueness of angular correlation functions is thus a check on the decay scheme and on the quality of the experimental work.

In this chapter, it will be shown that if reorientation is not a factor results of the radiothorium

correlation measurement cannot be interpreted in terms of a simple decay scheme which assigns spin zero to the ground states of radiothorium and ThX. Arguments will be advanced to show that reorientation is unlikely to be important, and that it is also unlikely that the correlation data can be convincingly explained even if the ground states are allowed spins different from zero. The most probable summary of the results therefore is that a choice must be made between believing this experiment or believing that radiothorium has a decay scheme with only two gamma rays.

Section 5.2. The Possibility of Reorientation

If the magnetic moment of the excited ThX nucleus formed by alpha emission is subjected to a magnetic field, it will classically execute a precessional motion about the field direction. The angular momentum vector will therefore, in general, have a variable component along the direction of alpha emission which will be called the Z axis. Quantum mechanically the effect is described by saying there is a probability of executing transitions from the original m state to 2J other m states.

Equation 2.1 for the angular correlation holds only if there is negligible probability of a change in m be-

fore the gamma ray is emitted. The possibility that the nucleus be exposed to a field arises from magnetic effects of the electronic structure belonging to the excited nucleus and to adjacent atoms as well if the nucleus is in a solid.

The question of whether reorientation is a worry involves the problem of estimating the relation of the lifetime for gamma emission to the Larmor precessional period for the ThX nucleus in the appropriate field. An upper limit on the gamma life of half a microsecond has been established experimentally and general lifetime formulations (5.2, 5.3) applied to the gamma rays in this experiment, (which are probably electric quadrupole) indicate the half life should even be ten to a hundred times smaller. The trouble comes in trying to estimate a field at the nucleus in order to get the Larmor frequency. If the radiothorium source were a gas there would surely be no reorientation problem because the electronic ground state of ThX has zero angular momentum so that there would be no electronic magnetic moment to interact with the nuclear moment. In a solid, electrons are shared by nuclei and the field at a nucleus can generally be expected to be zero even if the atoms of the solid have angular momentum when isolated in space. It is probably an additional help that the ThX atom which

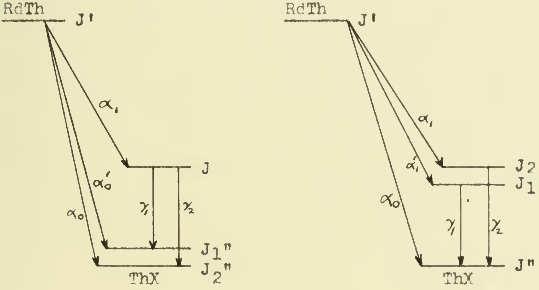
recoils through the atoms of the source and probably stops before the gamma ray is emitted has no angular momentum unless excited by collisions. The magnetic field that is effective in this problem is the root mean square field in a non-metallic solid. H. Primakoff has very kindly talked over the problem of estimating such a field and has ventured the opinion that the field would be incapable of producing reorientation in a time as long as a microsecond. He did point out that for intermediate cases between zero and full reorientation the angular correlation could not, in general, be expressed as a sum of the undisturbed correlation and an isotropic term.

In addition to the rather indefinite arguments against reorientation in the preceding paragraph, there is also one bit of experimental evidence that serves as a plausibility argument against reorientation. Namely, that a fairly sharp alpha-gamma correlation has been reported in the decay of ThC. The electronic configuration of the daughter in that case is $^2P_{\frac{1}{2}}$ and the energy of the emitted γ ray is even lower than in the present case.

Section 5.3. Possible Decay Schemes for Radiothorium

Before discussing the angular correlation data two decay schemes for RdTh will be presented as well as some evidence as to the nature of the gamma radiation.

Scheme (a), below, is consistent with all previous experiments and (b) violates only a contention by Rosenblum (5.4) that the longer range group of alpha rays was observed to be broadened by an amount consistent with the existence of two unresolved groups 2.5 kev apart.



Scheme (a) Scheme (b)

γ_1 is the 83.3 kev radiation and γ_2 the 86.8 kev gamma.

It should be stated that there seems no particular experimental reason to doubt Rosenblum. The 40 kev alpha fine structure of ThC has been resolved by a number of observers in the past twenty years and it is plausible that

a separation ten percent this size could be noticed as a broadening. Rosenblum's experience with alpha fine structure is unrivaled as B.B. Kinsey has remarked in discussion of this point.*

There is strong evidence that γ_1 and γ_2 are electric quadrupole radiations. That they are of the same type comes from the fact that the ratio of conversion electrons to gamma rays, computed from Surugue and Tsien's electron counting and Riou's gamma ray work, is the same for both. Riou's figure of 0.025 gamma rays per disintegration used with the knowledge that 28% of the alpha particles leave the ThX nucleus excited gives them a conversion coefficient of ten. Conversion takes place in the L, M, and higher shells since K conversion is energetically impossible. Fraser (5.5) has measured the L and M conversion coefficients of the 84 kev radiation of ${}_{70}Yb^{170}$ and their sum is 3.6. Extrapolation of Z to 88 gives a figure like ten. The argument is completed by the fact that the transition in Yb can be identified as electric quadrupole from other considerations.** The conclusion can be checked by use of recent calculations of the L conversion coefficient in heavy elements (5.6).

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* Private communication.

** M. Goldhaber, private communication.

The identification of γ_1 and γ_2 as quadrupole or higher from the presence of terms in $\cos^4\theta$ in their angular correlations can be used with the experimental upper limit on their lives of 0.5×10^{-6} sec. and with theoretical estimates of gamma lifetimes to support the internal conversion evidence that they can only be electric quadrupole.

The table below shows the half lives predicted for 85 kev and Z = 88 by Weisskopf's recent theory (5.2) and by the more conventional Axel-Dancoff lifetime formulation (5.3).

	Weisskopf	Axel-Dancoff
Electric Dipole	1.4×10^{-13} sec.	1.8×10^{-13} sec.
Magnetic Dipole	2×10^{-10}	6.1×10^{-8}
Electric Quadrupole	3.8×10^{-7}	6.1×10^{-8}
Magnetic Quadrupole	5.6×10^{-4}	4.5×10^{-2}
Electric Octupole	1.7	4.5×10^{-2}

If these lifetimes are believed literally, the experimental knowledge that the half life is less than 0.5×10^{-6} sec. allows only electric or magnetic dipole and electric quadrupole. The correlation evidence rules out the dipoles. Actually the theories are guaranteed by their authors only to a factor of $10^{\pm 2}$. The leeway might barely allow magnetic quadrupole on the Weisskopf theory, however, the

conversion would be so high for magnetic quadrupole that the gamma rays could not have been observed in this experiment.

Further discussion will consider both gamma rays established as electric quadrupole.

<u>Section 5.4</u>. <u>Discussion of the Correlation Data</u>

The angular correlations to be explained were exhibited in Figs. 4.11 and 4.12 and are summarized in the table below.

Radiation	Energy	$W(\theta)$	Probable error
γ_1	83.3 kev	$W_1 = 1+6.81\cos^2\theta-7.22\cos^4\theta$	8%
γ_2	86.8	$W_2 = 1+7.2\cos^2\theta-6.2\cos^4\theta$	$\sim$ 30%

Because W_2 is poorly known it will not be used as a basis for involved argument. The discussion will assume no reorientation and show the difficulties of trying to fit W_1 to schemes (a) or (b) with $J' = 0$. Use will be made of the boundary condition provided by the emission of the 36.8 kev radiation, γ_2, to the ground state of ThX and this state will also be assigned spin zero.

Consider scheme (a) first and express relative parities with + or - according to whether they are even or odd with respect to the ground state of ThX. The E^2 (electric

quadrupole) nature of γ_2 establishes J as 2^+; that of γ_1 allows only $J_1^\pi = 0,1,2,3,4$ all +. The α_1 group can only have L = 2 which fixes the parity of the ground state of RdTh as +. The α_0' transition therefore proceeds between states of the same parity and so must have even L. This restricts the possible values of J_1^π to 0,2,4. $J_1^\pi = 4$ is highly unlikely because the large angular momentum change for α_0' would be expected to make that group so weak relative to α_0 that the main group of alpha particles would not appear broadened.

At this point the angular correlation W_1 should be invoked to decide which of the possible spins 0,2,4 is correct. The theoretical correlations for scheme (a) and those spins are plotted in Figs. 5.0, 5.1 and 5.4. Their equations are listed below:

Spin Sequence	$L_\alpha : L_\gamma$	F(θ) (No special normalization)
0-2-0	2:2	$F_0 = \cos^2\theta - \cos^4\theta$
0-2-2	2:2	$F_2 = 5+9\cos^2\theta - 12\cos^4\theta$
0-2-2	2:2,1	$F_2' = 5+9\cos^2\theta-12\cos^4\theta+ \frac{21}{5}\beta^2(1+\cos^2\theta) - 2\sqrt{\frac{21}{5}}\,\beta\cos\delta(3\cos^2\theta-1).$
0-2-4	2:2	$F_4 = 15 + 6\cos^2\theta - \cos^4\theta$

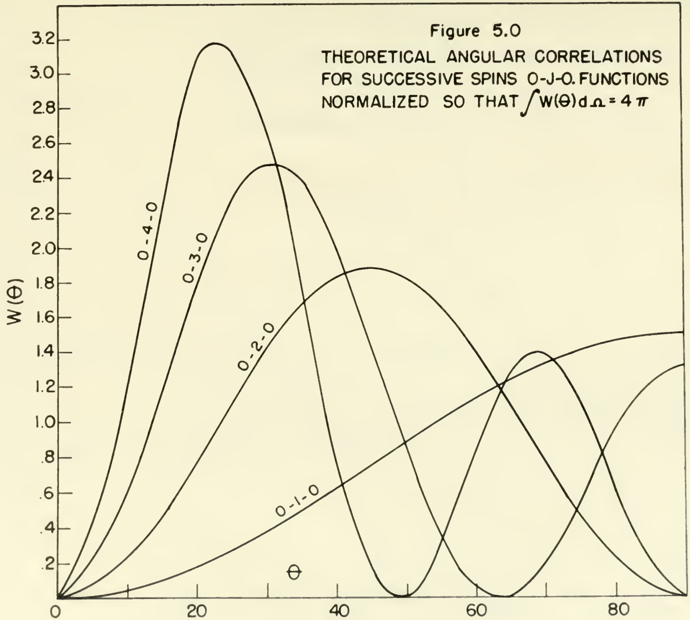


Figure 5.0
THEORETICAL ANGULAR CORRELATIONS FOR SUCCESSIVE SPINS 0-J-0. FUNCTIONS NORMALIZED SO THAT $\int W(\Theta)\, d\Omega = 4\pi$

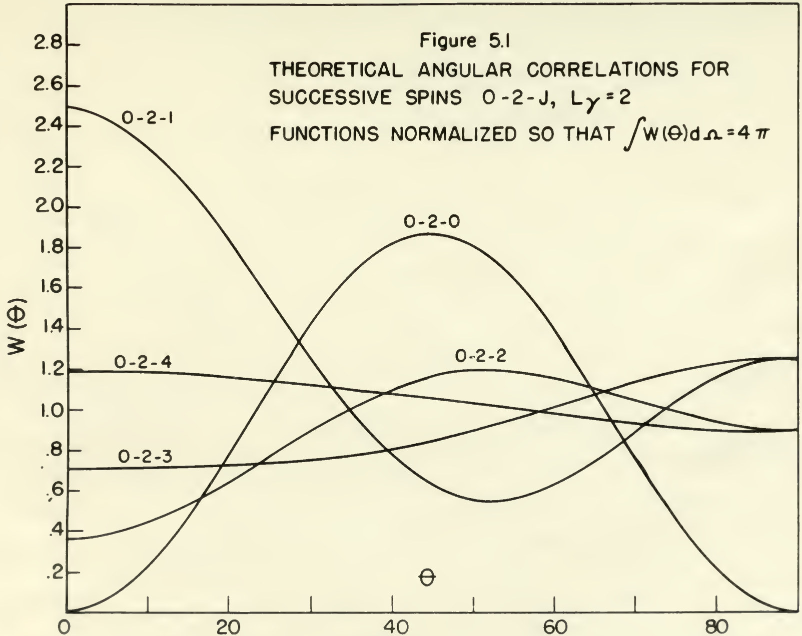


Figure 5.1

THEORETICAL ANGULAR CORRELATIONS FOR SUCCESSIVE SPINS 0-2-J, $L_\gamma = 2$

FUNCTIONS NORMALIZED SO THAT $\int W(\Theta)\, d\Omega = 4\pi$

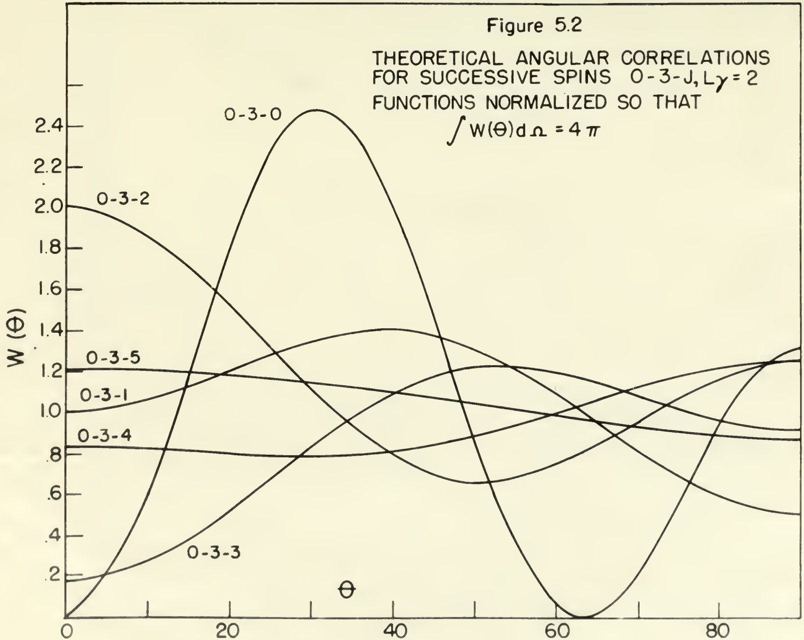


Figure 5.2

THEORETICAL ANGULAR CORRELATIONS FOR SUCCESSIVE SPINS 0-3-J, $L_\gamma = 2$ FUNCTIONS NORMALIZED SO THAT $\int W(\Theta)\, d\Omega = 4\pi$

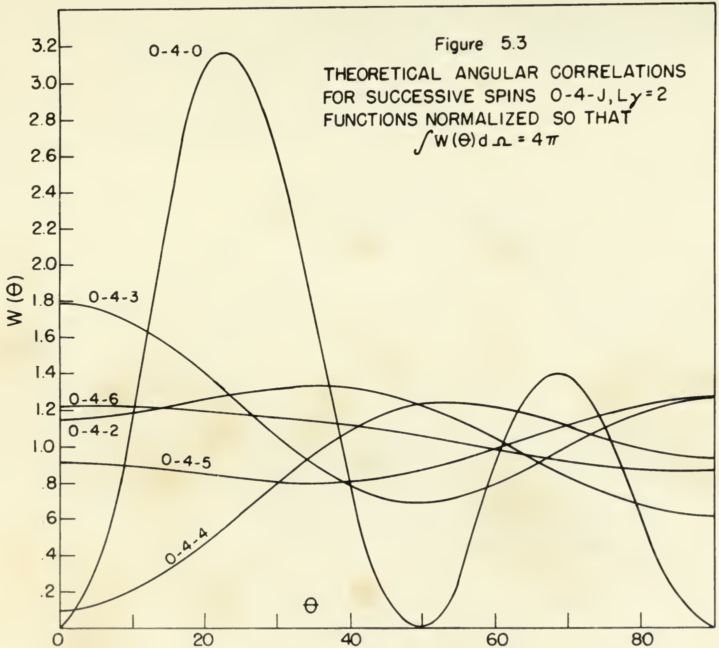


Figure 5.3

THEORETICAL ANGULAR CORRELATIONS FOR SUCCESSIVE SPINS 0-4-J, $L_\gamma = 2$ FUNCTIONS NORMALIZED SO THAT

$$\int W(\Theta)\, d\Omega = 4\pi$$

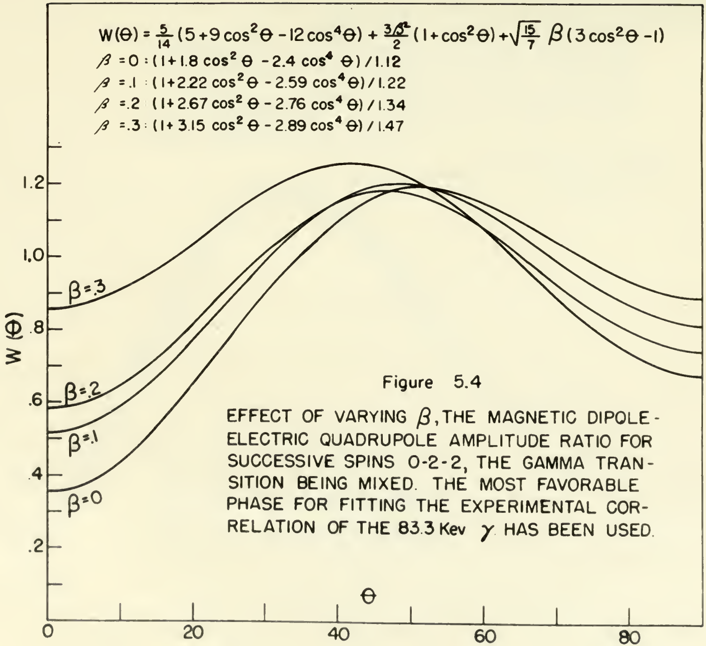


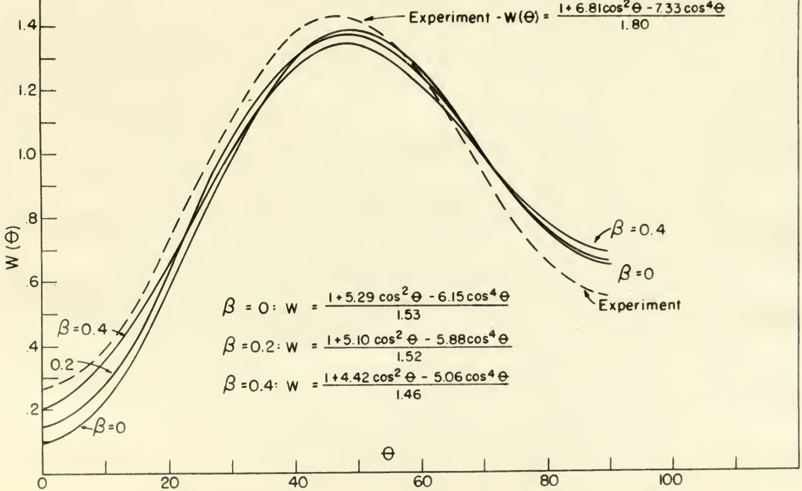
Figure 5.4

EFFECT OF VARYING β, THE MAGNETIC DIPOLE-ELECTRIC QUADRUPOLE AMPLITUDE RATIO FOR SUCCESSIVE SPINS 0-2-2, THE GAMMA TRANSITION BEING MIXED. THE MOST FAVORABLE PHASE FOR FITTING THE EXPERIMENTAL CORRELATION OF THE 83.3 Kev γ HAS BEEN USED.

Figure 5.5

ATTEMPT TO FIT THE EXPERIMENTAL CORRELATION OF THE 83.3 Kev GAMMA RAY WITH A MIXTURE FOR BOTH TRANSITIONS

The scheme assumed for the calculation is 2-3-2 with $L_\alpha = 2,4$; $L_\gamma = 2,1$. The alpha amplitude ratio = 0.3 and the dipole to quadrupole amplitude ratio is called β.



The angular correlation, F_2', takes account of the possibility of magnetic dipole as well as electric quadrupole radiation. For this case β is the dipole-quadrupole amplitude ratio and δ the corresponding phase difference.

Comparison of the equation for W_1 with those of the theoretical correlations or comparison of Fig. 4.11 with the corresponding graphs shows no possibility of agreement.

The next step is to try the alternative decay scheme, (b). Since γ_1 and γ_2 are E_2, $J_1 = J_2 = 2^+$. The fact that J_2 and J_1 have the same parity excludes an electtic dipole transition of 3.5 kev between them. The other radiations allowed by selection rules can be dismissed as factors which might influence the angular correlation because of very long lifetimes resulting from the low energy. Decay scheme (b) should therefore yield two angular correlations of $\cos^2\theta - \cos^4\theta = \sin^2 2\theta$. These are definitely not observed.

The only way that W_1 could be reconciled with scheme (a) with $J' = J_2'' = 0$ would be to assume that J_1'' was also zero and reorientation produced a strong assymetry in the original symmetric function $\sin^2 2\theta$. It would be necessary to have $J_1'' = 0$ because only zero of the three possible values $J_1'' = 0, 2, 4$ gives a correlation function which is sharper than the observed

function, W_1, and could be distorted into W_1 by reorientation. An additional very implausible assumption of a strong difference in the effect of reorientation on the correlations of γ_1 and γ_2 would have to be made to explain how the two original correlations, both $\sin^2 2\theta$, became altered into the distinctly different functions W_1 and W_2. The same assumption would be necessary to explain W_1 and W_2 for scheme (b) in terms of ground states with zero spin.

If either of the simple schemes (a) or (b) is to fit the experimental angular correlations without reorientation it will be necessary to relax the condition that both the ground state of radiothorium and the ground state of ThX have no spin. When this is done the number of possible values of J', J, J" are limited only by the fact that the spins must be relatively small integers and J and J" may not differ by more than two. It becomes out of the question to calculate all the possible angular correlations, especially since mixtures must generally be considered for both alpha particles and gamma rays. The search for a fit to W_1 for ground states with spin can therefore not be considered concluded until theorems are developed which state the restrictions imposed on the angular momenta entering into the correlation calculation by the requirements that $W(\theta)$ have the general characteristics

of W_1. Nevertheless there is a strong feeling after calculating a number of mixtures that the observed correlation W_1 is a sharper function than any theoretical function with $L_\gamma \leq 2$ except $\sin^2 2\theta$. Figures 5.0-5.4 show a number of correlation functions which, while only one is a mixture, should illustrate how hard it is to find as sharp a function as W_1 without exceeding $L_\gamma = 2$. Equations of such of these correlations as have $L_\gamma \leq 2$ as well as of a number of others are tabulated in Appendix 2. Figure 5.5 shows the theoretical correlation which came closest to W_1. This was a mixture of alphas and gammas and the spin sequence was 2-3-2. The fit is not good and its forced nature is evident.

It is worth mentioning that the observed correlation, W_1, can be fitted very well by a normalized addition of 38% $\sin^2 2\theta$ and 62% $5+9\cos^2\theta-12\cos^4\theta$. The latter function is the correlation for quadrupole gamma radiation and successive spins 0-2-2. Decay scheme (a) could be modified to include this spin sequence without contradicting experimental evidence by the addition of a level with spin two very close to the 3.5 kev level which would be assigned the spin $J_1'' = 0$. It is quite possibly true that W_1 cannot be made to fit the sum of $\sin^2 2\theta$ and any correlation function having L = 1,2 and spins 0-J-J" regardless of the values allowed J and J". Despite the fact that the fit to 0-2-2

may even be unique it is not convincing and its implications will not be pursued for it was obtained by an ad hoc assumption of an extremely unlikely character.

Section 5.5. Summary

The present experimental work has confirmed all the main features of earlier studies on the decay scheme of radiothorium in an independent way. In addition, a pronounced angular correlation between alpha radiation and a gamma ray which can be identified with one of 83.3 kev, previously reported, has been measured fairly accurately. It has not proved possible to fit the correlation to a decay scheme involving only the four levels known to exist. This suggests the possibilities, (a) there is appreciable reorientation of the excited ThX nucleus before gamma emission, (b) there is a large systematic error in the angular correlation measurement, and (c) that the radiothorium decay scheme, while mainly established, is not correct in detail.

In regard to the decay scheme, there seems no reason to ask more information from alpha fine structure experiments. However, the gamma radiation cannot be considered to have been studied extensively by techniques having high energy resolution. Analysis of the gamma rays with a crystal spectrometer of excellent resolution* would

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* See 5.7 for description of a measurement of an 80 kev gamma ray energy to an accuracy of better than 0.01% by means of a crystal spectrometer.

be desirable even aside from the possibility that it might validate the results of the present experiment and such an investigation is recommended.

The fact that there does not appear to be a theoretical correlation function besides $\sin^2 2\theta$, with $L_\gamma \leq 2$ and with the sharply varying character of the observed angular correlation, W_1, suggests that the spin sequence 0-2-0 plays a prominent role in the radiothorium decay scheme. There is consequently no reason to suspect that the ground states of RdTh and ThX do not have zero spin. The present experiment will stand as strong evidence that both ground states are indeed spinless, if theorems on alpha-gamma correlations can be developed to support the suspicion that, even allowing mixed radiations, W_1 is sharper than all correlations with dipole and quadrupole gammas except the correlation for spins 0-2-0. A theoretical study of angular correlations between alpha and gamma rays with a view to seeing whether such theorems exist is therefore recommended.

CHAPTER VI

Possible Alpha-Gamma Angular Correlation Measurements

There are a large number of alpha-gamma angular correlations which might be measured. The naturally radioactive actinium series, for example, has numerous gamma rays in cascade with alpha particles.* Many of the possible correlation experiments would require special instrumentation. However, the equipment built for the radiothorium measurement can be applied with only small modification to several other correlations. It seems worthwhile to enumerate these briefly.

In the thorium series ThX could be measured provided that the 238 kev gamma ray to be expected from the 4.6% fine structure, (6.2), is not highly converted. The growth of spurious coincidences after separation of ThX from its decay products might be a more serious problem than the build-up in the RdTh experiment and it might be necessary to break the decay chain at thoron.

The recent correlation measurement, (6.3) in the ThC alpha decay could profitably be repeated because there is suspicion that alpha particles from ThC' were allowed to contribute to the coincidences counted.

* See 6.1 for decay schemes and references.

A gamma-alpha correlation could be measured between the 2.2 mev gamma ray and the 8.78 mev alpha particle from ThC'. The existence of a correlation would be interesting because of the 3.10^{-7} sec. half life of ThC' and the fact that the electronic configuration has an angular momentum of two. Scattering of energetic gamma rays in the radiothorium correlation apparatus should be investigated before undertaking this measurement. The corresponding experiment on RaC' which lives 500 times longer might be interesting.

The uranium series has two elements whose decays should allow possible correlation experiments with the radiothorium equipment. Ionium emits quanta of 68 and 190 kev in 0.5% and 0.3% of its disintegrations. Although these gammas are of weak intensity and the specific activity of ionium is low the experiment should not be hard. This is because radium is the immediate decay product and its long half life eliminates build-up after separation. Differential discrimination on the alpha pulses would be unnecessary so that thick sources could be used.

The disintegration of radium produces a single gamma ray of 184 kev in about one percent of the decays. The low gamma intensity might make it advisable to remove

radon formed during the run as a measure against build-up. Techniques for doing this are well established.

The lifetimes of the gamma rays from the decays of ionium and radium can be expected to be short enough to give negligible loss of coincidences with a resolving time of $.3 \times 10^{-6}$ sec. If the lives were too long to give coincidences, it is probable the gamma rays would be so highly converted they would not have been observed. For these elements, as well as ThX, reorientation should not be more serious than in the radiothorium correlation measurement because the normal electronic structures of the product nuclei are without angular momentum.

APPENDIX I

Correction for Counter Size

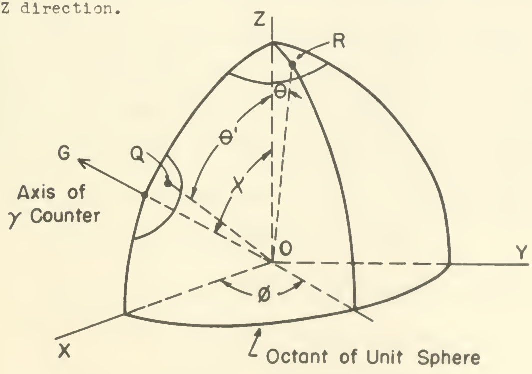
Let the counters be circular and move on the surface of the unit sphere centered on the point source. The alpha counter is to subtend an angle 2Δ and the gamma counter angle will be 2δ. The angle between counters is called χ.

If $W(\theta) = \lambda+\mu\cos^2\theta+\nu\cos^4\theta$ the relative probability of gamma emission into unit solid angle at angle θ to the direction of the alpha ray is found by normalization to be

$$p(\theta) = \frac{\lambda+\mu\cos^2\theta + \nu\cos^4\theta}{4\pi\left(\lambda+\frac{\mu}{3}+\frac{\nu}{5}\right)}$$

It is desired to find $P(\chi)$ the relative probability that the alpha and gamma rays from a particular disintegration enter their respective counters.

The figure shows the placement of the detectors and the notation used. The axis of the alpha counter defines the Z direction.



Q is a point on the gamma counter with coordinates θ,φ, relative to the axis of that counter as pole. The point R is on the alpha counter and has coordinates θ,φ. Let $\theta'' =$ Angle GOR.

The probability that the gamma ray would be sent into its counter if the alpha particle was emitted along Z is then,

$$p'(\chi) = \int_{\theta_1=0}^{\delta} \int_{\phi_1=0}^{2\pi} \frac{\lambda + \mu\cos^2\theta' + \nu\cos^4\theta'}{4\pi(\lambda + \frac{\mu}{3} + \frac{\nu}{5})} \sin\theta_1 d\theta_1 d\phi_1$$

With $\cos\theta' = \cos\chi\cos\theta_1 - \sin\chi\sin\theta_1\cos\phi_1$.

Substituting for $\cos\theta'$ and working out the integrals gives,

$$\iint \sin\theta_1 d\theta_1 d\phi_1 = 2\pi(1-\cos\delta).$$

$$\iint \cos^2\theta' \sin\theta_1 d\theta_1 d\phi_1 = \pi\left[1-\cos\delta - \tfrac{1}{3}(1-\cos^3\delta)\right] + \pi\cos\delta\sin^2\delta\cos^2\chi.$$

$$\iint \cos^4\theta' \sin\theta_1 d\theta_1 d\phi_1 = \tfrac{3}{4}\pi\left[1-\cos\delta - \tfrac{2}{3}(1-\cos^3\delta) + \tfrac{1}{5}(1-\cos^5\delta)\right]$$
$$+ \tfrac{3}{2}\pi\cos\delta\sin^4\delta\cos^2\chi - \tfrac{3}{4}\pi\cos\delta\sin^2\delta(1-\tfrac{7}{3}\cos^2\delta)\cos^4\chi.$$

The same results would have been obtained if the expression for $\cos\theta'$ had contained a positive instead of a negative sign which may easily be verified. Substitution yields,

$$2(\lambda + \tfrac{\mu}{3} + \tfrac{\nu}{5})p' = \lambda(1-\cos\delta) + \tfrac{\mu}{2}\left[1-\cos\delta - \tfrac{1}{3}(1-\cos^3\delta)\right]$$
$$+ \tfrac{3}{8}\nu\left[1-\cos\delta - \tfrac{2}{3}(1-\cos^3\delta) + \tfrac{1}{5}(1-\cos^5\delta)\right]$$
$$+ \tfrac{1}{2}\cos\delta\sin^2\delta(\mu + \tfrac{3}{2}\nu\sin^2\delta)\cos^2\chi - \tfrac{3}{8}\nu\cos\delta\sin^2\delta(1-\tfrac{7}{3}\cos^2\delta)\cos^4\chi.$$

The relative probability wanted is given by,

$$P(\chi) = \int_{\theta=0}^{\Delta}\int_{\phi=0}^{2\pi} p'(\theta'') \frac{\sin\theta d\theta d\phi}{4\pi}.$$

Where,

$$2(\lambda + \tfrac{\mu}{3} + \tfrac{\nu}{5})p'(\theta'') = a + b\cos^2\theta'' + c\cos^4\theta''$$

and

$$a = \lambda(1-\cos\delta) + \tfrac{\mu}{2}\left[1-\cos\delta - \tfrac{1}{3}(1-\cos^3\delta)\right] + \tfrac{3}{8}\nu\left[1-\cos\delta - \tfrac{2}{3}(1-\cos^3\delta) + \tfrac{1}{5}(1-\cos^5\delta)\right]$$

$$b = \tfrac{1}{2}\cos\delta\sin^2\delta(\mu + \tfrac{3}{2}\nu\sin^2\delta)$$

$$c = -\tfrac{3}{8}\nu\cos\delta\sin^2\delta(1-7/3\cos^2\delta)$$

so that,

$$8\pi(\lambda + \tfrac{\mu}{3} + \tfrac{\nu}{5})P = a\iint\sin\theta d\theta d\varphi + \iint\cos^2\theta''\sin\theta d\theta d\varphi + c\iint\cos^4\theta''\sin\theta d\theta d\varphi$$

With $\cos\theta'' = \cos\chi\cos\theta + \sin\chi\sin\theta\cos\varphi$.

The integrals have the values of the previous three if Δ is written instead of δ. Thus, $P(\chi)$ may be written down from the result for $p'(\chi)$ with λ, μ, ν replaced respectively by a, b, c.

$$\begin{aligned}4(\lambda + \tfrac{\mu}{3} + \tfrac{\nu}{5})P(\chi) &= a(1-\cos\Delta) + \tfrac{b}{2}\left[1-\cos\Delta - \tfrac{1}{3}(1-\cos^3\Delta)\right] \\ &+ \tfrac{3}{8}c\left[1-\cos\Delta - \tfrac{2}{3}(1-\cos^3\Delta) + \tfrac{1}{5}(1-\cos^5\Delta)\right] \\ &+ \tfrac{1}{2}\cos\Delta\sin^2\Delta(b + \tfrac{3}{2}c\sin^2\Delta)\cos^2\chi - \tfrac{3}{8}c\cos\Delta\sin^2\Delta(1 - \tfrac{7}{3}\cos^2\Delta)\cos^4\chi\end{aligned}$$

Replacing a, b, and c by their values above and collecting terms gives the desired probability:

$$\begin{aligned}P(\chi) = \frac{(1-\cos\delta)(1-\cos\Delta)}{4(\lambda + \frac{\mu}{3} + \frac{\nu}{5})}\Big\{&\lambda + \tfrac{\mu}{12}\left[4 - \cos\delta\cos\Delta(1+\cos\delta)(1+\cos\Delta)\right] \\ &+ \tfrac{\nu}{40}\Big[(1-\cos\delta)^2(8+9\cos\delta+3\cos^2\delta) + 5\sin^2\delta\cos\delta(1+\cos\delta)(2+\cos\Delta)(1-\cos\Delta) \\ &- \tfrac{1}{8}\cos\delta(1+\cos\delta)(1-7/3\cos^2\delta)(1-\cos\Delta)^2(8+9\cos\Delta+3\cos2\Delta)\Big] \\ &+ \tfrac{1}{4}\cos\delta(1+\cos\delta)\cos\Delta(1+\cos\Delta)\left[\mu + \tfrac{3}{8}\nu(1+3\cos^2\delta+3\cos^2\Delta-7\cos^2\delta\cos^2\Delta\right]\cos^2\chi \\ &+ \tfrac{9}{64}\nu\cos\delta(1+\cos\delta)\cos\Delta(1+\cos\Delta)(1-\tfrac{7}{3}\cos^2\delta)(1-\tfrac{7}{3}\cos^2\Delta)\cos^4\chi\Big\}\end{aligned}$$

The quantity in braces is defined as $f(\chi)$ mentioned in Chapter III.

In this experiment $\Delta = 10°$ and $\delta = 11°$. With these values

$$f(\chi) = \lambda + 0.0165\mu + 0.602 \times 10^{-3}\nu + (0.951\mu + 0.0921\nu)\cos^2\chi + 0.843 \cos^4\chi.$$

For the sharp correlation, $W(\theta) = \cos^2\theta - \cos^4\theta$,

$$f(\chi) = 0.0159 + 0.859\cos^2\chi - 0.843 \cos^4\chi.$$

APPENDIX 2

Alpha-Gamma Angular Correlation Functions

A number of alpha-gamma angular correlation functions have been worked out and are listed below. Simple correlations have been normalized so that $W(\frac{\pi}{2}) = 1$ except for $\sin^2 2\theta$. No particular normalization has been used for mixtures. The small Greek letters which appear in $W(\theta)$ are amplitude ratios, or phases if they are used as angles. For mixed alpha transitions amplitudes and phases are relative to the lowest listed alpha angular momentum. Their meaning should be apparent.

Correlations where the gamma ray is not a single multipole are mixtures of magnetic dipole and electric quadrupole radiation with β and Δ representing respectively the dipole to quadrupole amplitude ratio and phase.

I. SIMPLE CORRELATIONS

	J'	J	J"	λ	μ	ν
(a) $L_\alpha = 0,\ L_\gamma = 1$						
	0	1	0	1	-1	
	0	1	1	1	1	
	0	1	2	1	-1/7	
	1	1	1	1	-1/3	
(b) $L_\alpha = 1,\ L_\gamma = 2$						
	0	1	1	1	1	
	0	1	2	1	-3/5	
	0	1	3	1	3/13	
(c) $L_\alpha = 2,\ L_\gamma = 1$						
	0	2	1	1	-3/5	
	0	2	2	1	1	
	0	2	3	1	-1/5	
	2	1	2	1	7/91	
(d) $L_\alpha = 2,\ L_\gamma = 2$						
	0	2	0	0	1	-1
	0	2	1	1	-3	4
	0	2	2	1	9/5	-12/5
	0	2	3	1	-6/7	3/7
	2	1	2	1	3/7	0
	2	2	2	1	45/73	-48/73
	2	3	2	1	144/41	-165/41
(e) $L_\alpha = 3,\ L_\gamma = 1$						
	0	3	2	1	-1/2	
	0	3	3	1	1	
	0	3	4	1	-3/13	
(f) $L_\alpha = 3,\ L_\gamma = 2$						
	0	3	1	1	6	-5
	0	3	2	1	-12/5	3
	0	3	3	1	21/11	-30/11
	0	3	4	1	-1	2/3
	0	3	5	1	12/23	-3/23

SIMPLE CORRELATIONS (con'd)

J'	J	J''	λ	μ	ν
(g) $L_\alpha = 4,\ L_\gamma = 2$					
0	4	2	1	60/17	-45/17
0	4	3	1	-15/7	18/7
0	4	4	1	37/19	-54/19
0	4	5	1	-12/11	9/11
0	4	6	1	30/49	-9/49
(h) $L_\alpha = 5,\ L_\gamma = 2$					
0	5	3	1	30/11	-21/11
0	5	4	1	-2	7/3
0	5	5	1	57/29	-84/29
0	5	6	1	-15/13	12/13
0	5	7	1	15/22	-5/22
(i) $L_\alpha = 6,\ L_\gamma = 2$					
0	6	5	1	-21/11	24/11
0	6	6	1	81/41	-120/41

The foregoing tabulation may be extended somewhat by use of the property of $W(\theta)$ that if $L_\alpha = L_\gamma$, $W(\theta)$ is the same for the sequence J''-J-J' as for J'-J-J''. For example, $W(\theta)$ for $L_\alpha = L_\gamma = 2$ and successive spins 1-2-0 is found from the correlation for 0-2-1, $L_\alpha = L_\gamma = 2$, to be $1-3\cos^2\theta + 4\cos^4\theta$.

II. MIXTURES

J'	J	J"	L_α	L_γ	
(a) Alpha Mixed-Gamma Simple					
1	2	0	1,3	2	$W=3(1+\cos^2\theta)+2\alpha^2(1+6\cos^2\theta-5\cos^4\theta)+2\sqrt{6}\,\alpha\cos\delta\,(1-9\cos^2\theta+10\cos^4\theta)$
1	2	1	1,3	1	$W=141(1-\frac{21}{47}\cos^2\theta)+144\alpha^2(1-\frac{1}{2}\cos^2\theta)-6\sqrt{5}\,\alpha\cos\delta\,(1-3\cos^2\theta)$
1	2	1	1,3	2	$W=7(1+\frac{3}{7}\cos^2\theta)+8\alpha^2(1-\cos^2\theta+\frac{5}{3}\cos^4\theta)+2\sqrt{6}\,\alpha\cos\delta\,(-1+11\cos^2\theta-\frac{40}{3}\cos^4\theta)$
1	3	1	2,4	2	$W=2(27+78\cos^2\theta-55\cos^4\theta)+3\alpha^2(17+60\cos^2\theta-45\cos^4\theta)$ $+\frac{12}{3}\alpha\cos\delta\,(3-24\cos^2\theta+25\cos^4\theta)$
2	2	0	1,3	2	$W=5-3\cos^2\theta+\alpha^2(5-12\cos^2\theta+15\cos^4\theta)+4\alpha\cos\delta\,(3\cos^2\theta-5\cos^4\theta)$
2	2	0	0,2,4	2	$W=4+\frac{5}{7}\alpha^2(5+9\cos^2\theta-12\cos^4\theta)+\frac{1}{7}\beta^2(17+60\cos^2\theta-45\cos^4\theta)$ $+4\sqrt{5/14}\,\alpha\cos\Delta_2(1-3\cos^2\theta)+\frac{4}{\sqrt{14}}\beta\cos\Delta_4(-3+30\cos^2\theta-35\cos^4\theta)$ $+\frac{4\sqrt{5}}{7}\alpha\beta\cos(\Delta_4-\Delta_2)(3-24\cos^2\theta+25\cos^4\theta)$
2	3	1	2,4	2	$W=3(31-42\cos^2\theta+55\cos^4\theta)+15\alpha^2(5+3\cos^4\theta)+4\sqrt{10}\,\alpha\cos\delta\,(3-21\cos^2\theta+20\cos^4\theta)$
2	3	2	2,4	2	$W=41+144\cos^2\theta-165\cos^4\theta+5\alpha^2(10+9\cos^2\theta-9\cos^4\theta)+2\sqrt{10}\,\alpha\cos\delta\,(-3+33\cos^2\theta-40\cos^4\theta)$
2	4	2	3,5	2	$W=30+3\cos^2\theta+13\cos^4\theta+\alpha^2(27+24\cos^2\theta-7\cos^4\theta)+2\sqrt{2}\,\alpha\cos\delta\,(3-21\cos^2\theta+20\cos^4\theta)$
(b) Alpha Simple Gamma Mixed					
0	2	2	2	1,2	$W=\frac{5}{14}(5+9\cos^2\theta-12\cos^4\theta)+\frac{3}{2}\beta^2(1+\cos^2\theta)-\sqrt{\frac{15}{7}}\,\beta\cos\Delta\,(3\cos^2\theta-1)$
(c) Alpha and Gamma Mixed					
2	3	2	2,4	1,2	$W=\frac{5}{4}(41+144\cos^2\theta-165\cos^4\theta)+\frac{25}{4}\alpha^2(10+9\cos^2\theta-9\cos^4\theta)$ $+5\sqrt{\frac{5}{2}}\,\alpha\cos\delta\,(-3+33\cos^2\theta-40\cos^4\theta)+\beta^2(73-9\cos^2\theta)$ $+\frac{5}{2}\alpha^2\beta^2(31-9\cos^2\theta)+3\sqrt{10}\,\alpha\beta^2\cos\delta\,(-1+3\cos^2\theta)+3\sqrt{30}\,\beta\cos\Delta(1-3\cos^2\theta)$ $+25\sqrt{\frac{15}{8}}\,\alpha^2\beta\cos\Delta(1-3\cos^2\theta)+30\sqrt{3}\,\alpha\beta\cos\delta\cos\Delta(3\cos^2\theta-1)$

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